

Article

Fly Ash-Based Geopolymer Composites: A Review of the Compressive Strength and Microstructure Analysis

Shaker Qaidi ^{1,2,*}, Hadee Mohammed Najm ^{3,*}, Suhad M. Abed ⁴, Hemn U. Ahmed ⁵, Husam Al Dughaihi ⁶, Jawad Al Lawati ⁶, Mohanad Muayad Sabri ⁷, Fadi Alkhatib ⁸ and Abdalrhman Milad ^{6,*}

¹ Department of Civil Engineering, College of Engineering, University of Duhok, Duhok 42001, Iraq

² Department of Civil Engineering, College of Engineering, Nawroz University, Duhok 42001, Iraq

³ Department of Civil Engineering, Zakir Husain Engineering College, Aligarh Muslim University, Aligarh 202002, India

⁴ Department of Highways & Airports Engineering, College of Engineering, University of Diyala, Diyala 32001, Iraq

⁵ Civil Engineering Department, College of Engineering, University of Sulaimani, Sulaimaniyah 16278, Iraq

⁶ Department of Civil and Environmental Engineering, College of Engineering, University of Nizwa, Nizwa P C 616, Ad-Dakhiliyah P.O. Box 33, Oman

⁷ Peter the Great St. Petersburg Polytechnic University, 195251 St. Petersburg, Russia

⁸ Department of Structural Engineering, Faculty of Civil Engineering and Built Environment, Universiti Tun Hussein Onn Malaysia (UTHM), Batu Pahat 86400, Malaysia

* Correspondence: shaker.abdal@uod.ac (S.Q.); gk4071@myamu.ac.in (H.M.N.); a.milad@unizwa.edu.om (A.M.)



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Abstract: Geopolymer (GP) concrete is a novel construction material that can be used in place of traditional Portland cement (PC) concrete to reduce greenhouse gas emissions and effectively manage industrial waste. Fly ash (FA) has long been utilized as a key constituent in GPs, and GP technology provides an environmentally benign alternative to FA utilization. As a result, a thorough examination of GP concrete manufactured using FA as a precursor (FA-GP concrete) and employed as a replacement for conventional concrete has become crucial. According to the findings of current investigations, FA-GP concrete has equal or superior mechanical and physical characteristics compared to PC concrete. This article reviews the clean production, mix design, compressive strength (CS), and microstructure (Ms) analyses of the FA-GP concrete to collect and publish the most recent information and data on FA-GP concrete. In addition, this paper shall attempt to develop a comprehensive database based on the previous research study that expounds on the impact of substantial aspects such as physio-chemical characteristics of precursors, mixes, curing, additives, and chemical activation on the CS of FA-GP concrete. The purpose of this work is to give viewers a greater knowledge of the consequences and uses of using FA as a precursor to making effective GP concrete.

Keywords: geopolymer; fly ash; carbon dioxide emission; microstructure; alumino-silicates; compressive strength; mix design

1. Introduction

The most utilized construction material is concrete because of its performance, cost, ability to form different shapes, and availability of raw materials locally [1–3]. However, the increasing demand for concrete for various construction applications has led to a corresponding high consumption of raw materials utilized in the manufacture of concrete [1,4,5]. The production of PC, which is the primary binder in concrete, has also been shown to have a hugely detrimental influence on the environment. It has been estimated that approximately one ton of CO₂ is emitted into the environment for every ton of PC produced [6–9]. It has been estimated that between 2017 to 2050, the manufacture of PC annually will grow by 50%. The substantial rise in the demand for PC to produce concrete

is expected to substantially increase the amount of CO₂ emitted into the environment. CO₂ emissions estimated between 85 to 105 gigatons could be emitted into the atmosphere over the next 33 years because of the production of PC [6,10,11]. In addition to the high emission linked with PC production, about 2 to 3% of the worldwide energy demand is also consumed [6,11]. Figure 1 shows the global cement production and the amount of pollution associated with it. These high CO₂ emissions ensuing from PC production would hinder the world from achieving its sustainability goals and result in more detrimental impacts on the environment. Thus, this study aims to explore alternative sustainable materials such as GP concrete that can be used in place of PC concrete [2,12].

GP concrete is deemed the next generation of concrete because of its sustainability benefits as it offers an innovative way to totally eliminate PC as a binder in the manufacture of concrete, thereby resulting in a substantial reduction in CO₂ emissions [13–15]. Generally, in the literature, ligands with precursors based on alumina and silica are treated as GPs, while alkali-activated cement is based on precursors rich in calcium. In contrast to PC concrete, the binder phase in GP concrete is formed because of the process known as geopolymerization. Geopolymerization occurs when silicate and aluminate monomers in alumino-silicate precursors are activated with an alkali/acidic medium [16]. The alumino-silicate precursors used in GPs can be natural materials (e.g., kaolin, mining waste) or waste materials (e.g., palm oil ash, FA, rice husk ash, granulated blast slag) [17]. Figure 2 presents the common alumino-silicate precursors used in GP concrete. In addition to the sustainability benefits of GP concrete [18–20], the use of GP concrete also opens a pathway to effectively manage wastes that could have an adverse effect on health, safety, and the environment [21]. Studies have also shown that GP concrete with a similar cost to that of PC concrete can be produced while reducing CO₂ emission by about 22–72% [22]. In addition, GP concrete has been shown to have superior mechanical characteristics [23–26], an adjustable thermal expansion coefficient [27], high-temperature resistance [28], and acid resistance [29–32]. Figure 3 presents a schematic diagram of using GP concrete as a sustainable construction material.

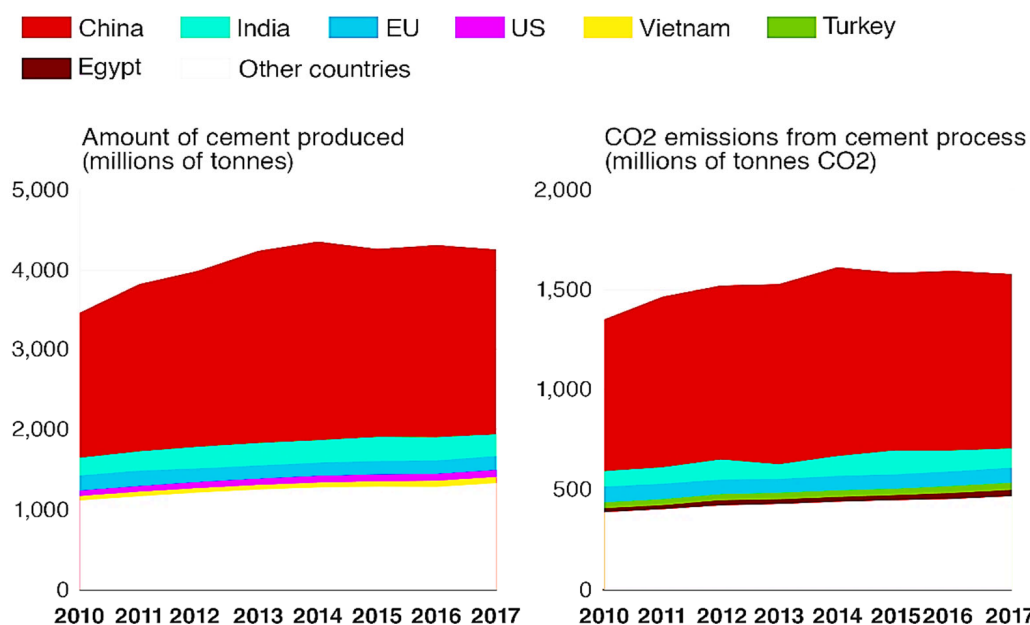


Figure 1. Global cement production volumes and process-related carbon dioxide emissions. Adapted from [33].

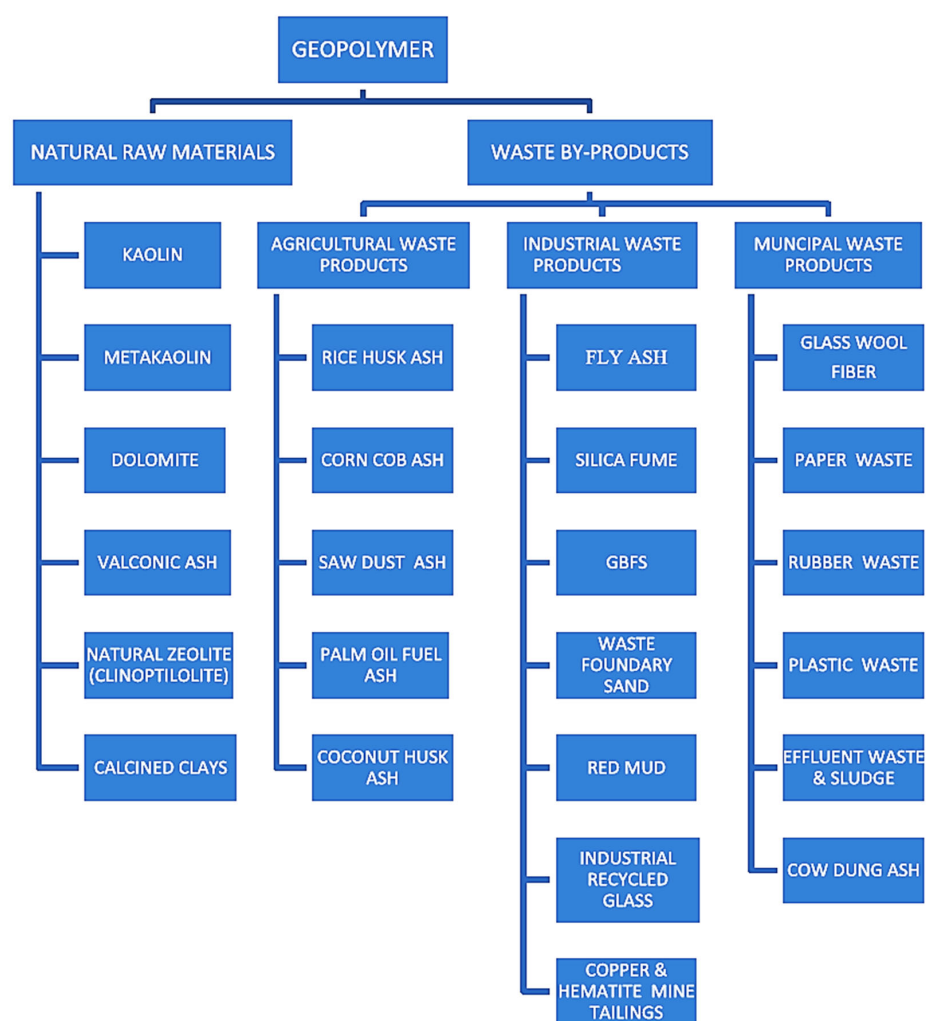


Figure 2. Precursor materials that are commonly utilized in GP composites. Adapted from [34].

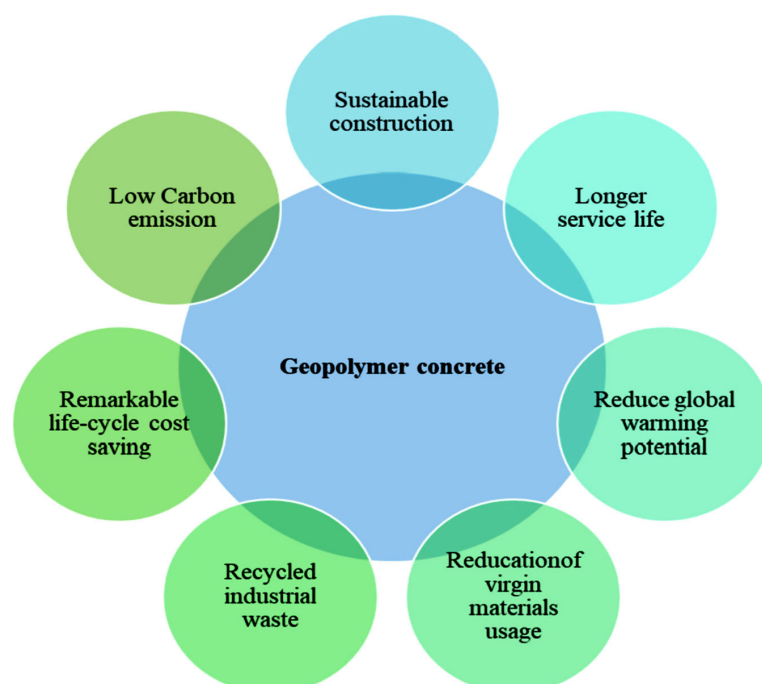


Figure 3. Benefits of GP concrete in sustainable construction. Adapted from [35].

GPs are composed of the alumino-silicate precursor which is composed of amorphous silica and alumina alongside an activator that acts as the dissolving agent. Geopolymerization is the process by which zeolitic-like materials are converted into a three-dimensional (3D) alumino-silicate gel [1,36–38]. A polycondensation procedure involving silicate and aluminate in an alkaline environment develops the corresponding performance of the generated binder [27]. The common activators used in synthesizing GPs are either MOH-type caustic alkalis or $R_2O \cdot (n)SiO_2$ -type silicates or both. M in the MOH indicates the alkali-metal such as K, Na, and Ca in the activator, which is generally sodium hydroxide (NaOH), sodium carbonate ($NaCO_3$), potassium hydroxide (KOH), or sodium sulfate (Na_2SO_4) [39]. In addition to the common activators mentioned, various alternative activators such as rice husk ash and silica fume have been utilized as activator components in synthesizing GPs. Compared to the hydration reaction of PC, the water used in the synthesis of GPs only helps in producing a workable mixture and does not participate in the geopolymerization and polycondensation process [40]. Figure 4 presents a schematic diagram of GP production [41–44].

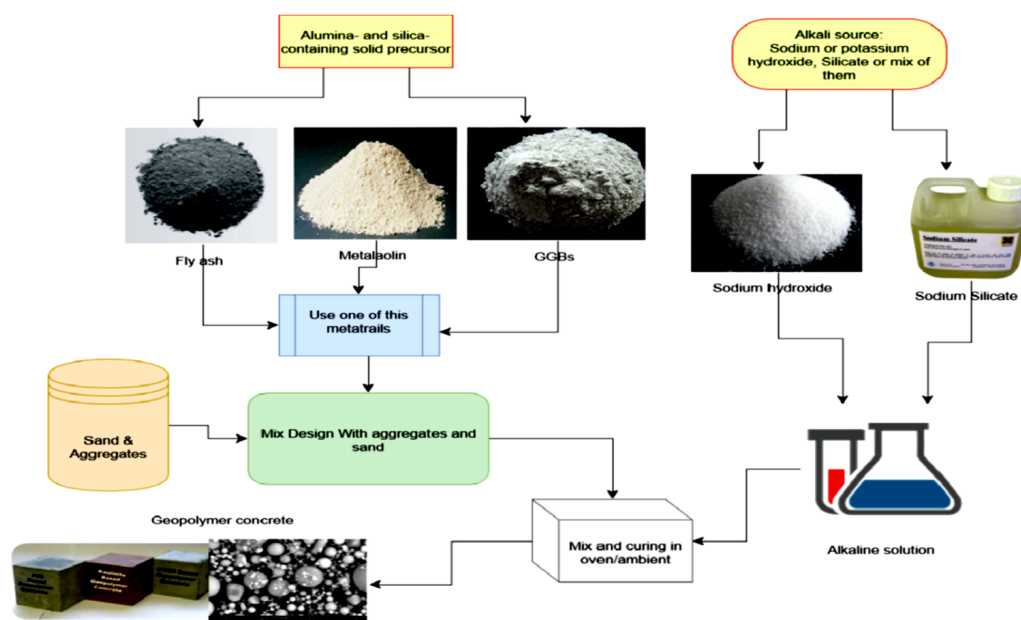


Figure 4. Production of GP. Adapted from [35].

This study aims to analyze and answer a lot of worries concerning the viability of FA-GP concrete as the next-generation sustainable concrete. Despite these numerous advantages, the impact of FA on the long-term characteristics of GP concrete remains a source of worry that must be fully understood before FA-GP concrete can be considered a sustainable alternative. In a summary, the purpose of this work is to give viewers a greater knowledge of the consequences and uses of using FA as a precursor to making effective GP concrete [45–47].

Depending on their local availability, use, and production cost, various types of precursors have been used to produce GPs [5,36,48]. The most common precursor used to make GP concrete is FA because of its high composition of silicate and aluminate coupled with its local availability. FA has been extensively used in PC concrete to partially replace PC as the binder [49]. Figure 5 presents many applications of FA compounds. The global FA generation is anticipated to reach over 363 million tons per year, with India leading the way, followed by China and the US [13,22]. Figure 6 presents global FA generation and usage. As the precursor, about 450 kg/m³ of FA is utilized for every ton of FA-GP concrete [22]. Because FA is the primary ingredient used for the synthesis of FA-GP concrete, it is anticipated that about 2 billion tons of FA would have been needed to substitute 100% PC [22,50]. As there is a substantial quantity of FA available worldwide,

its use in developing FA-GP concrete can be deemed a sustainable tactic [51]. This review paper focuses on the up-to-date development of FA-GP concrete. The emphasis is on identifying how the silicon/aluminum (S/A) ratio proportions, the additives, and the kind and quantity of alkali sol are employed to controllably enhance the qualities of FA-GP concrete. The critical property of concrete, namely the CS, is also discussed. A relationship between the Ms characteristics and CS of the FA-GP concrete is also established.

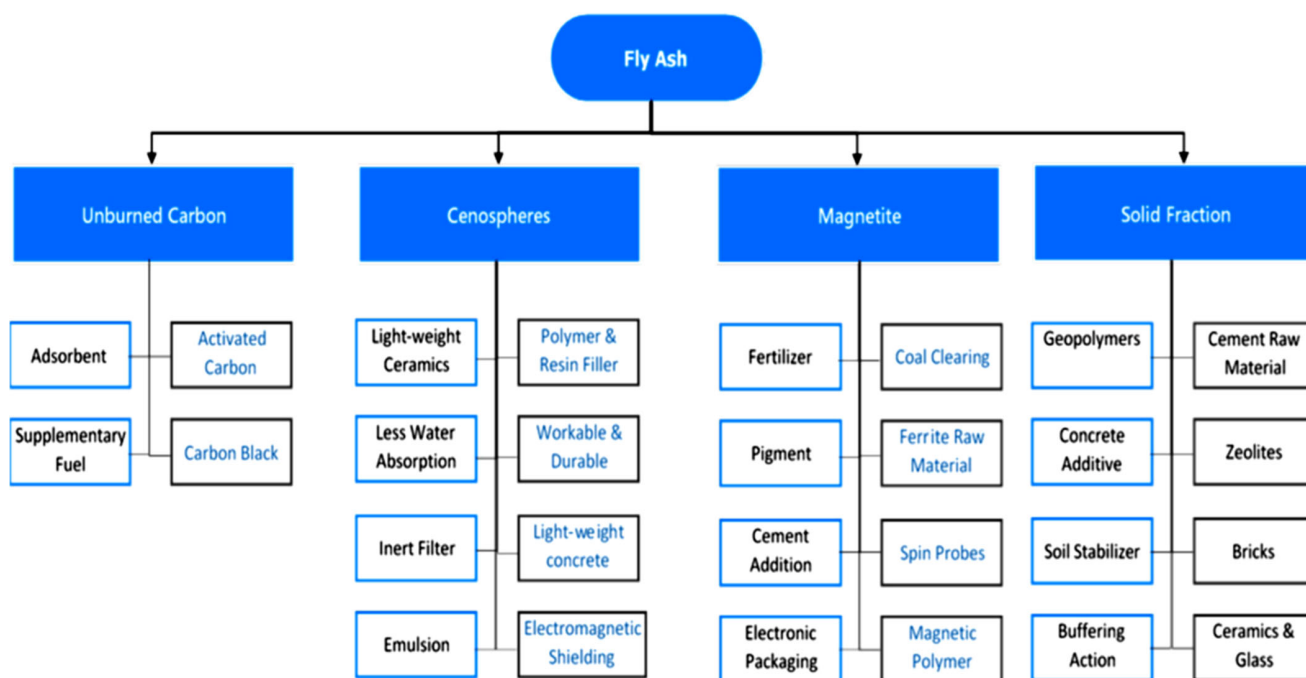


Figure 5. FA's main components and applications. Adapted from [52].

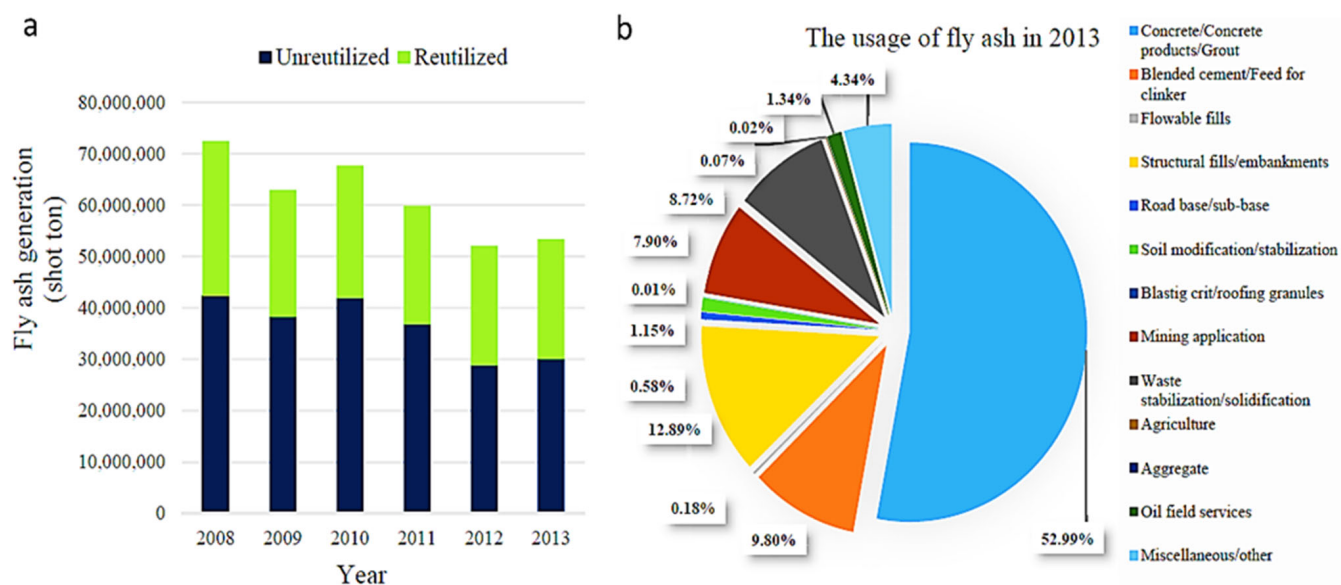


Figure 6. (a) FA generation in the United States from 2008 to 2019; (b) FA application fields in the United States in 2019. Adapted from [53].

2. Significance of Study

This study aims to analyze and answer a lot of worries concerning the viability of FA-GP concrete as the next-generation sustainable concrete. Numerous investigators that presented evaluations on FA-GP concretes discovered that the strength characteristics of the FA-GP concrete might be equal, if not considerably higher, compared to those of PC concrete [54,55]. Furthermore, the application of FA-GP concrete technologies may give environmental and economic benefits. As a result, FA-GP concrete is growing in recognition in the construction sector. Despite these numerous advantages, the impact of FA on the long-term characteristics of GP concrete remains a source of worry that must be fully understood before FA-GP concrete can be considered a sustainable alternative. A small number of review articles have been published that provide a complete overview of the uses and effects of FA on the different technical features of GP concrete. Similarly, the current work intends to evaluate and compile the outcomes of numerous investigations under one tent, with a focus on the Ms characteristics of FA-GP concrete. This review study combines the clean manufacturing of FA particles and parameters influencing the properties of FA-GP concrete and addresses the Ms characteristics and CS of FA-GP concrete in depth. In a summary, the purpose of this work is to give viewers a greater knowledge of the consequences and uses of using FA as a precursor to making effective GP concrete

3. Geopolymerization

Geopolymerization is a chemical reaction between alumino-silicate precursors and an alkaline component that results in a hardened product at room temperature or occasionally at an increased temperature [56]. Alkali-activated binders or alkali-activated materials are two terms for the same thing [57]. Materials rich in silica and alumina, such as ferrous slag, non-ferrous slags, calcined clay, and natural pozzolans, can be utilized as a source of alumino-silicate in geopolymerization [58–60]. The suggested structure of GP by Davidovits [56] is presented in Equation (1).



In Equation (1), M is a symbol of the alkali-metal cation, n is a symbol of the gradation of polycondensation, w is a symbol of the quantity of chemically attached water molecules, and z is a symbol of the silicon to an aluminum ratio. The GP structures are classified into three varieties based on the value of z: -Si-O-Al-O- (Gel 1, S/A = 1), -Si-O-Al-O-Si-O- (Gel 2, S/A = 2), and -Si-O-Al-O-Si-O-Si-O- (Gel 3, S/A = 3) [61–63]. Figure 7 illustrates these structures.

Figure 8 presents a rough idea of how a conceptual model of [64] works in the GP reaction. Dissolution of alumina and silica happens in GP reactions because of an alkaline environment composed of sodium and potassium hydroxides and silicates; the rearrangement and swapping of dissolved species results in GP gel formation [65]. Based on the chemical composition of the precursors, gel formation in GP reactions is classified into two types. Blast furnace slag (BFS) is a high-calcium precursor that creates the (C-A-S-H)-type of gel, whereas metakaolin (MK) and FA are low calcium precursors that make the N-A-S-(H) type of gel [64,65]. Strength is developed as the GP gel hardens.

In comparison, five distinct chemical processes exist for the GP, as illustrated in Figure 8b showing the Shi, Jiménez, and Palomo conceptual model [66–68]. When activated silico-aluminous cementitious materials react with alkaline activators, GPs are generated. According to this paradigm, polymerization processes are categorized into four groups: deconstruction, gel formation, polycondensation, and crystallization.

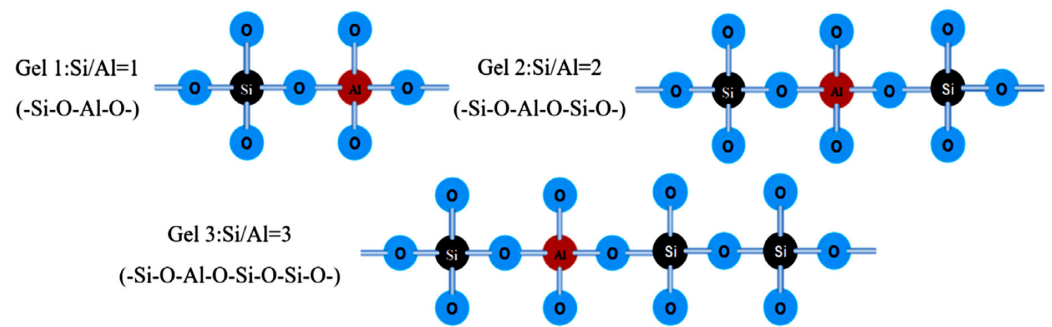


Figure 7. GP structures according to Davidovits [56]. Adapted from [66].

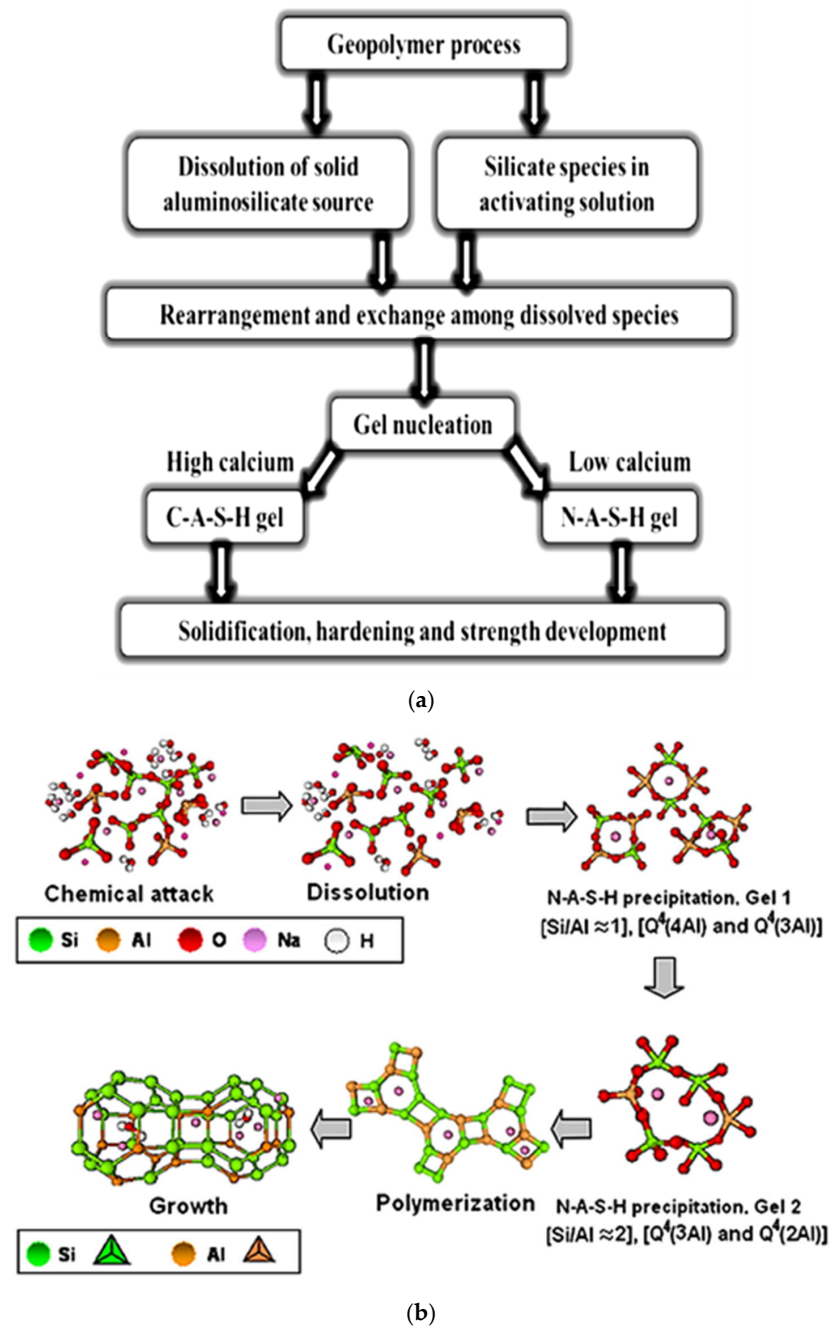


Figure 8. (a) A Provis conceptual model of geopolymerization [64,69]; (b) a Glukhovskiy conceptual model for geopolymerization [70].

4. FA-GP Concrete

FA is an abbreviation for “pulverized fuel ash” generated after the collection of boiler flue gas from coal power plants, as shown in Figure 9 [71,72]. The characteristics of the FA in terms of chemical and physical can be correlated to calcined condition and formation process. FA’s primary chemical compositions are SiO_2 , Al_2O_3 , and CaO [73], and the amorphous slope glass dosage has the greatest influence on activity [74–77]. The noxiousness of FA leachate exhibited negligible ecotoxicity [78]. SEM-EDS analysis indicates that FA possesses mullite and quartz phases [79] as shown in Figure 10. These phases are critical to the strength development of GPs [80–83].

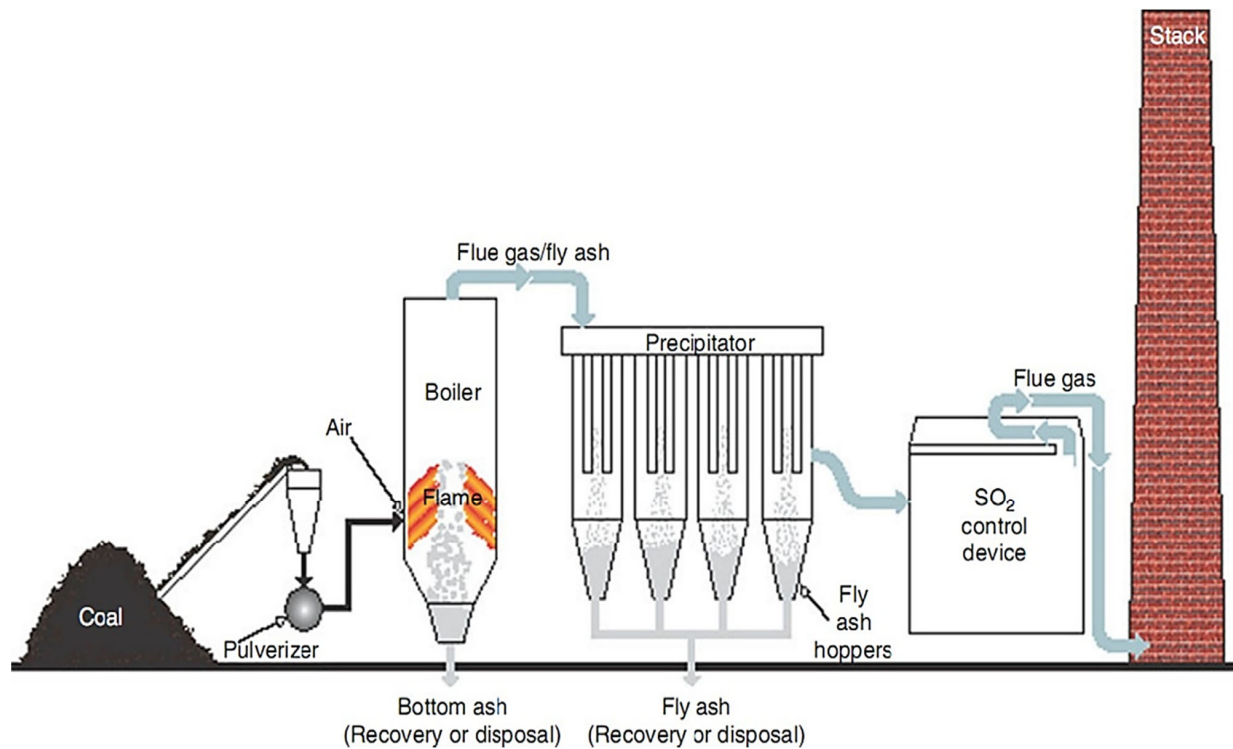


Figure 9. Clean production of FA. Adapted from [84].

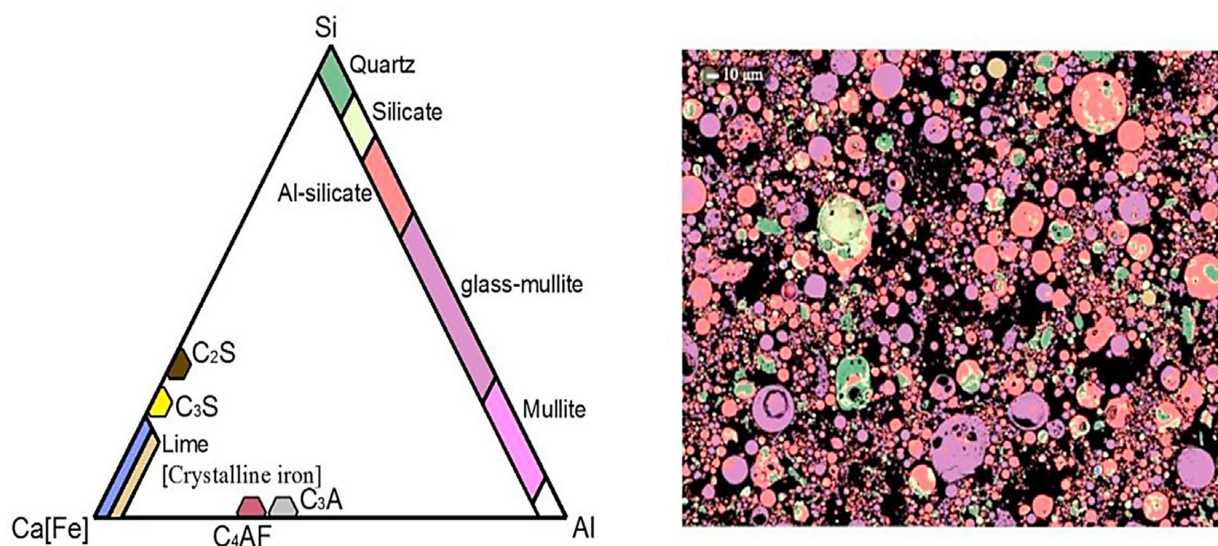


Figure 10. SEM-EDS characterization of FA mineral phase composites based on intuition. Adapted from [79].

The formation of FA-GP concrete is based on the breakdown of aluminosilicate in FA, which is accelerated by alkali, followed by polycondensation. Manufacturing is supposed to be more energy- and resource-efficient since reactions can occur at low temperatures. The actual responses that occur during the process, on the other hand, are exceedingly complicated and remain a mystery. Condensation between the resulting Si^{4+} and Al^{3+} molecules occurs because of FA-alkali reactions, followed by more intricate nucleation, oligomerization, and polymerization, culminating in a novel aluminosilicate-based polymer with a 3D network structure. In experiments or applications, the produced FA-GP concrete is placed in a mold and dried in an oven at a specific temperature or at room temperature for a specified period to form the structure indicated in Figure 11.

Alkali activation of FA is expected to be critical for geopolymerization formation: in an alkaline sol, the silica, alumina, or aluminosilicates in FA hydrolyze, and the -Si-O-Si- or -Si-O-Al- bonds of aluminosilicate break, releasing active Al^{3+} and Si^{4+} species, which combine to form nuclei and aluminosilicate oligomers composed of SiO_4 and AlO_4 tetrahedra. Based on the S/A ratio, the networks in aluminosilicate oligomers can be polyciliate -Al-O-Si-chain, polyciliate siloxo -Al-O-Si-Si-chain, or polyciliate disiloxo -Al-O-Si-Si-Si-. Al^{3+} partly replaces Si^{4+} in aluminosilicate monomers, and the resulting negative charge in the aluminosilicate bonds is neutralized by alkali cations such as K^+ and Na^+ [85,86], as illustrated in Figure 11. In this situation, the S/A ratio substantially impacts the final structure of the resulting GP composites [87]. It has been discovered, for example, that the S/A ratio in the FA reactor has a substantial influence on the pores (number and size) of crystalline products, which is one of the major characteristics governing the mechanical strength of GP compositions [88,89]. Table 1 outlines the various FA categories and mineral phases, providing details on their possible usage as GP precursors and the prospective end product.

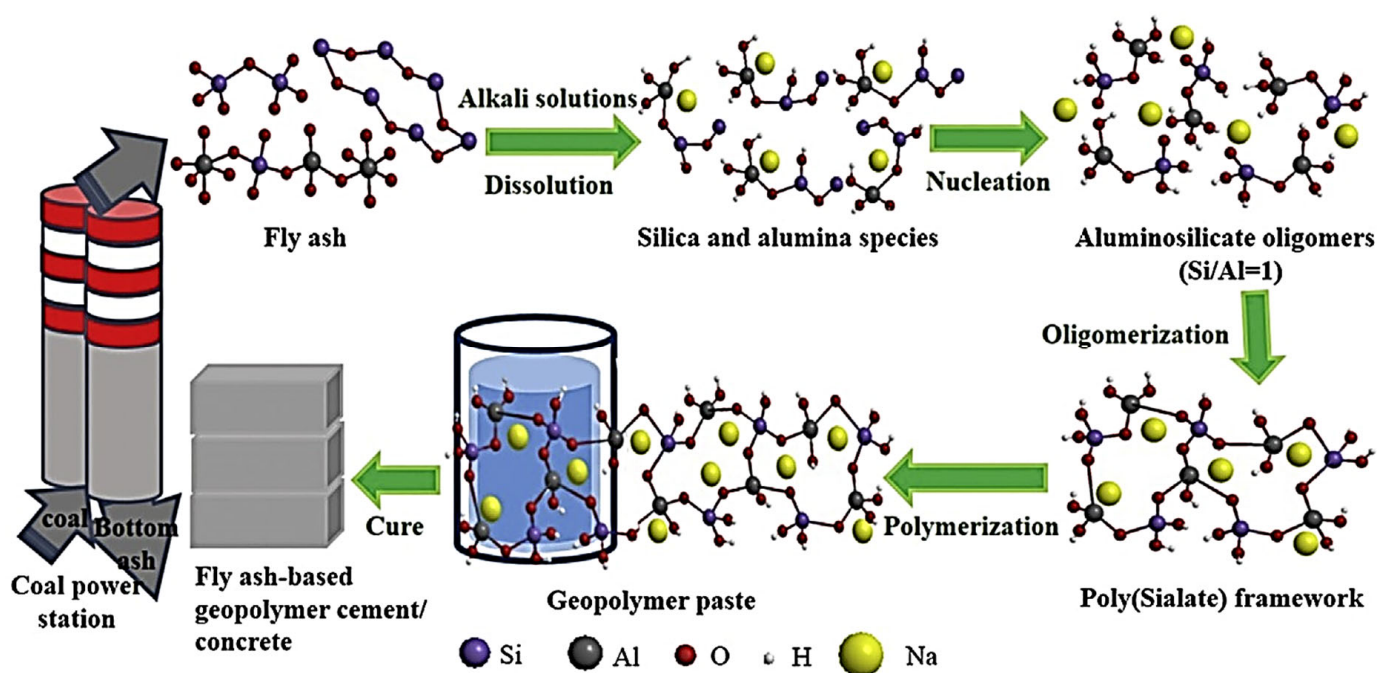


Figure 11. The schematic diagrams illustrate the pathway from FA to FA-GP concrete. Adapted from [90].

Table 1. FA classification and applications for the production of inorganic GPs. Adapted from [91].

FAs	Origin	Characteristic	Mineral Phases	Potential of Products	Potential for Geopolymerization
1. Pozzolanic (sialic)	Highly ranked fuels, bituminous with detrital quartz, kaolinite, illite, mica, k-feldspars	Maximum values of fine fraction, true density	Viscous melt (glass), minimum values of crystallization	Dense and porous GP pastes	Good activation but with a low rate of high dissolution pH of alkaline sol
2. Active low pozzolanic, calisialic, ferrisialic, ferricalisialic	Variable rank of fuel but mostly liquid with carbonates, sulfates, sulfides, detrital silicates	Maximum values of the water-soluble fraction	Oxyhydroxydes, sulfates, carb, and actives silicates	Porous matrix paste	Aam low alkaline sol and pH
3. Ferrisialic inert (sialic), calisialic	Variable rank of fuels, bituminous and sub-bituminous	High content of original infused quartz, max true density, min of water-soluble elements	Mullite and quartz	Dense and less porous composites	Low dissolution rate, high pH
4. Ferricalisialic	Low-rank fuels, lignites, bituminous gypsum, ferrite, calcite, dolomite	Max bulk density	Fe, Mg, Na phases	Porous product paste	High dissolution rate, poor polymerization
5. Sialic and calisialic mixed between inert and active	Lower-rank fuel, lignites and sub-bituminous with carbonate and sulfates	Maximum values of light fraction	Oxyhydroxides sulfates, carbonates, and active Ca and Caemg silicates	Paste and mortars, porous metallic corrosion, and pores	A medium reactive low alkaline sol

5. Chemical Composition of Binder Materials

Various parameters such as particle size, S/A ratio, amorphous composition, and phase composition affect the structure and characteristics of GPs [92]. Precursors such as FA have a high amount of alumina and silica, granting them the ability to act as supplementary cementitious materials. Silicon oxide and aluminum oxide are the two most common oxides found in FA. Because PC is made from limestone, it has more calcium oxide and silicon oxide but less aluminum oxide. Table 2 presents the conclusions of XRF experiments on FA industrial by-products utilized in the production of GPs that have been described in different articles. In addition, Figure 12 shows an XRD study of several raw materials, indicating the materials' phases. Figure 13 shows SEM of FA concrete in conjunction with EDS in the compositions of different hydration products in the same picture at multiple places.

Table 2. Oxide composition for PC and FA binder materials.

Material and Ref.	Composition (wt.%)															
	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	CaO	Pb	Na ₂ O	P ₂ O ₅	MnO	K ₂ O	SO ₃	MgO	SrO	TiO ₂	CuO	Cl	LOI
PC [93]	19.0	3.20	4.68	66.8	26.8	0.09	0.08	0.19	1.17	3.00	0.81	-	-	-	-	2.48
FA [94]	48.8	10.20	27.00	6.20	86.0	0.37	1.20	0.15	0.85	0.22	1.40	0.16	1.30	-	-	1.70
FA [95]	61.8	4.11	28.05	0.87	94.0	0.40	-	-	0.82	1.32	0.38	-	-	-	-	0.49

Table 2. Cont.

Material and Ref.	Composition (wt.%)															
	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	CaO	Pb	Na ₂ O	P ₂ O ₅	MnO	K ₂ O	SO ₃	MgO	SrO	TiO ₂	CuO	Cl	LOI
FA [96]	49.0	3.00	31.00	5.00	83.0	4.00	1.00	–	1.00	0.00	3.00	–	2.00	–	–	0.00
FA [93]	50.7	8.80	28.80	2.38	88.3	0.84	–	–	2.40	0.30	1.39	–	–	–	–	3.79
FA [97]	26.4	30.13	9.25	21.6	65.7	–	0.67	0.27	2.58	1.30	–	–	3.07	–	–	3.02
FA [98]	53.5	7.47	28.80	1.55	89.7	–	–	–	–	0.14	0.81	–	–	–	–	3.11
FA [99]	64.9	5.69	26.64	0.33	97.3	0.49	–	–	0.25	0.33	0.85	–	–	–	–	0.45
FA [100]	17.5	12.43	36.37	10.5	66.3	–	–	–	1.77	1.39	3.05	–	0.88	–	–	1.19
FA [101]	54.7	5.15	27.28	5.31	87.1	0.43	1.12	0.10	1.00	1.01	1.10	0.36	1.82	0.01	–	6.80
FA [102]	27.3	2.01	50.85	5.41	80.2	0.04	–	0.02	0.33	–	0.28	–	2.12	–	–	7.74
FA [103]	50.6	6.35	18.96	14.1	75.9	0.69	–	–	–	0.74	3.12	–	–	–	–	0.17
FA [71]	57.6	5.80	28.90	0.20	92.3	–	–	–	0.90	0.20	0.90	–	–	–	–	3.60
FA [104]	63.1	3.07	24.88	2.58	91.0	0.71	0.17	0.05	2.01	0.18	0.61	–	0.96	–	–	1.45
FA [105]	66.5	3.54	22.47	1.64	92.5	0.58	–	–	1.75	0.10	0.65	–	0.88	–	–	1.66
FA [106]	51.1	12.50	25.70	4.30	89.3	0.80	0.90	0.20	0.70	0.20	1.50	–	1.30	–	–	0.60
FA [107]	47.8	14.09	28.00	3.81	89.9	0.41	1.81	0.21	0.62	0.27	0.93	–	1.99	–	–	0.43
FA [108]	54.4	8.14	27.72	1.29	90.3	0.67	–	–	–	0.11	–	–	–	–	–	4.11
FA [109]	52.7	5.92	18.05	12.9	76.7	1.11	–	0.14	2.09	1.76	3.86	–	1.01	–	–	1.60
FA [110]	58.4	4.19	23.80	7.32	86.3	1.43	–	–	1.02	0.44	1.11	–	–	–	–	0.50
FA [111]	35.8	17.31	15.05	17.1	68.2	1.58	0.30	–	3.12	5.94	2.34	–	–	–	–	0.10
FA [112]	50.8	6.82	23.15	6.87	80.8	1.29	1.14	–	2.14	1.24	1.70	0.19	1.01	–	–	0.55

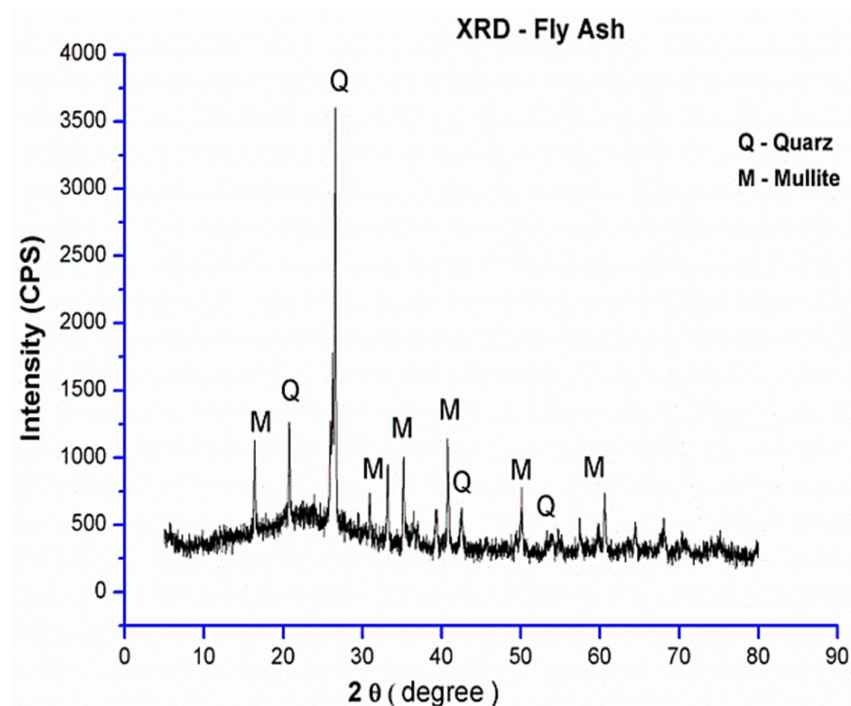


Figure 12. XRD of FA. Adapted from [113].

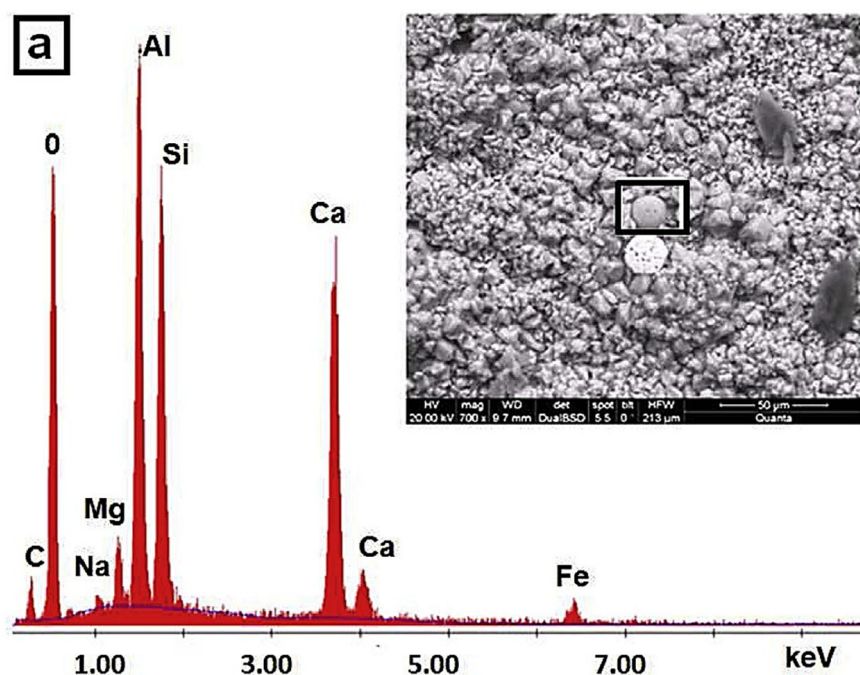


Figure 13. SEM and EDX curve showing the chemical composition of FA spots. Adapted from [49].

6. Mix Design of GP Concrete

GP concrete's CS is heavily influenced by its content and mix design. As demonstrated in Table 3, variables such as $\text{SiO}_2/\text{Al}_2\text{O}_3$, temperature curing, supplementary water, the molarity of $\text{Na}_2\text{O}/\text{Al}_2\text{O}_3$, and NaOH ratio have a critical influence on the CS of FA-GP concrete [114–116]. Pimraksa, Chindaprasirt, Rungchet, Sagoe-Crentsil, and Sato [117] observed that increasing the molarity of NaOH accelerates the geopolymerization process, leading to increased CS [118–121]. Moreover, the findings by Al Bakri, Kamarudin, Bnhussain, Nizar, Rafiza, and Zarina [122] and Songpiriyakij, Kubprasit, Jaturapitakkul, and Chindaprasirt [123] demonstrated that the $\text{Na}_2\text{SO}_3/\text{NaOH}$ ratio has a substantial impact on GP concrete strength growth. Other studies [124–133] have also observed that temperature and curing time have a considerable impact on the mechanical characteristics of GP concrete.

The creation of a standard for GP concrete is currently in its early stages. June [134], Ferdous, Manalo, Khennane, and Kayali [135] and Anuradha, Sreevidya, Venkatasubramani, and Rangan [136] offered numerous strategies for achieving mixed GP concrete proportions. According to Lokuge, Wilson, Gunasekara, Law, and Setunge [137], in comparison to PC concrete, creating the design mix of GP concrete is complicated and challenging as a result of the involvement of various variables.

The low-calcium FA was used in all experimental trials indicated in Table 3. The Fe_2O_3 percentage is around 10 to 20% by weight, while the CaO percentage is less than 5% [95,112,138]. According to research on the particle size distribution of FA, 80% of the particles of FA were smaller than 50 μm [127].

As in PC concrete, fine and coarse aggregates can be used as aggregates in GP concrete [126,127]. The alkali activator (AA) used to produce FA-GP concrete can be a combination of NaOH sol and Na_2SiO_3 sol. Aluminum oxides and silicon contribute to over 80% of the total weight, with a silicon-to-aluminum ratio of almost 2 for low-calcium FA. The binder is the primary distinction between GP concrete and PC concrete. The GP paste is made by reacting Al_2O_3 and SiO_2 within FA with an alkaline activator sol, which bonds unreacted components to the GP concrete [27]. Sand and gravels, as in PC concrete, make up roughly 75 to 80% of the GP concrete mass. The technologies now available for PC concrete can compute this component of GP concrete mixes [139]. Phoo-ngernkham, Phi-

angphimai, Damrongwiriyanupap, Hanjitsuwan, Thumrongvut, and Chindaprasirt [140] proposed using the multivariate adaptive regression spline (MARS) approach to formulate a GP concrete combination with a 28-day goal strength. This approach is based on contour plots generated between a variety of variables that affect GP concrete's CS. The method illustrated in Figure 14 can be utilized to build a GP concrete mix design.

The amounts and characteristics of the basic ingredients used in the creation of GP concrete influence its workability and CS. According to Pimraksa, Chindaprasirt, Rungchet, Sagoe-Crentsil, and Sato [117], increasing the dosage of NaOH sol increased the CS of GP concrete. It was also discovered that increasing the ratio of Na_2SiO_3 sol to NaOH sol by mass results in higher CS of GP concrete and that increasing the workability of GP concrete by incorporating superplasticizer, up to 4% FA by mass, improves the workability of GP concrete; however, this may hurt the CS of GP concrete. It was also discovered that as GP concrete's water content increases, its workability increases.

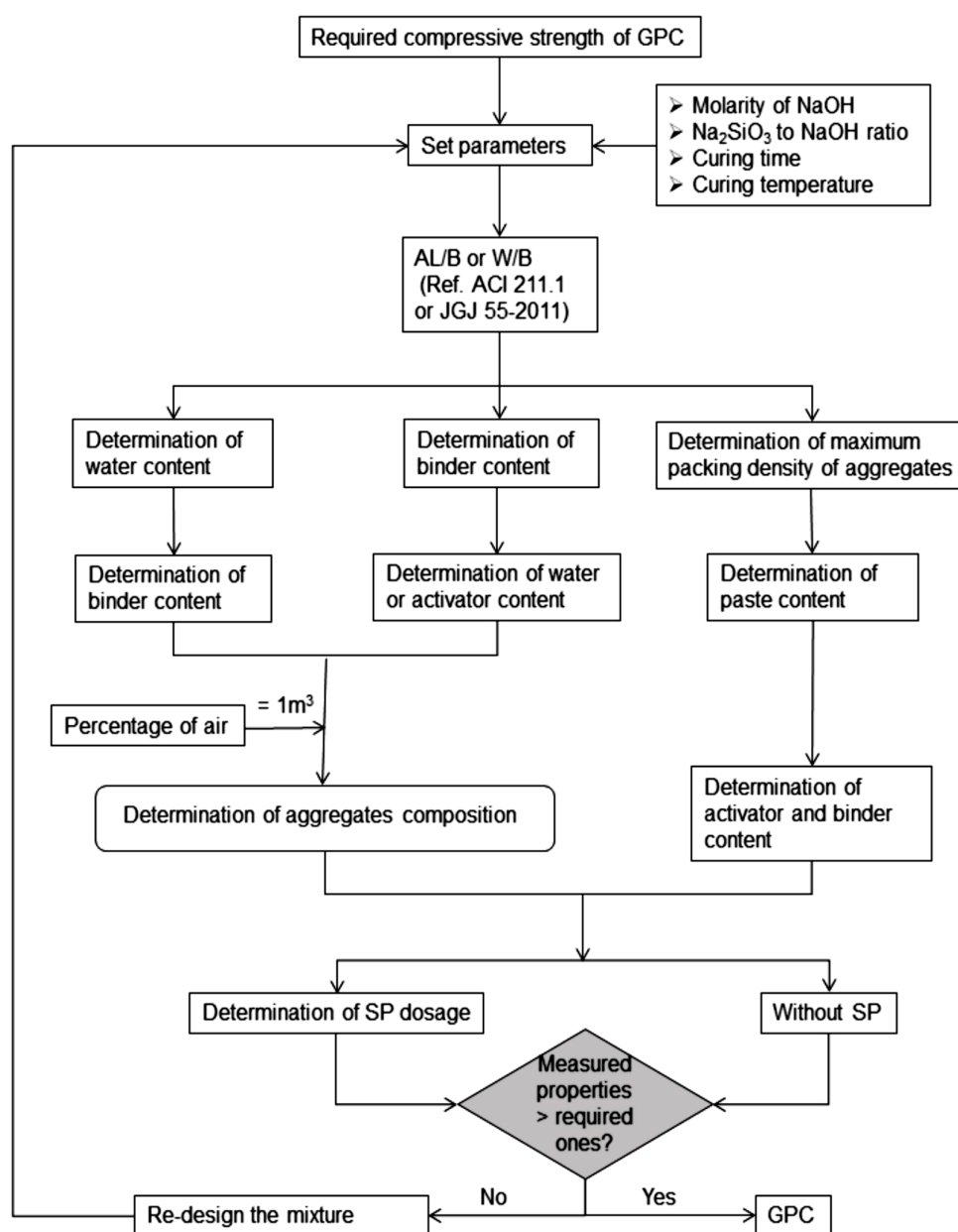


Figure 14. Mixture design method for GP concrete. Adapted from [141].

Table 3. Composition of FA-GP concrete.

Ref.	FA (Kg/m ³)	Aggregates (kg/m ³)		Alkaline Activator (AA) (kg/m ³)		NaOH Molarity (M)	Added Water (kg/m ³)	Curing Conditions		Strength (MPa)
		Fine	Coarse	Na ₂ SiO ₃ (SS)	NaOH (SH)			Temperature (C)	Time (h)	
[142]	450	500	1150	162	108	16	24	24	24	38.4
	450	500	1150	162	108	12	24	24	24	41.4
	450	500	1150	162	108	8	24	24	24	40.0
[143]	400	651	1209	114.3	45.7	14	22	22	24	26.7
[144]	349	620	1221		194	12	13.3	23	24	41.7
[145]	225	627	1164	112.5	45	14	24	24	24	41.1
[146]	400	548	1164	160	80		24	24	24	47.0
	300	623	1323	108	72		24	24	24	39.0
[147]	400	554	1293	113	45	14	100	100	24	33.5
[148]	400	850	950	143	57.0	12	40	70	24	53.5
	400	850	950	143	57.0	12	48	60	48	44.8
	400	850	950	143	57.0	12	60	70	24	37.3
	400	850	950	143	57.0	12	80	70	24	22.6
	400	850	950	143	57.0	12	80	70	24	22.6
[90]	400	658	1222	84	56.0	14	0.0	20–23		27.0
	400	658	1222	100	40.0	14	0.0	20–23	72	25.0
[124]	408	647	1201	103	41.0	14	0.0	90	24	89.0
	408	554	1294	103	41.0	14	21.3	30	24	32.0
[142]	476	554	1294	120	48.0	14	0.0	60	24	68.0
	408	647	1201	103	41.0	8.0	0.0	60	24	63.0
	476	554	1294	120	48.0	8.0	0.0	60	24	57.0
	408	647	1201	103	55.4	8.0	0.0	75	24	44.0
	408	647	1201	103	41.0	10	7.5	60	24	45.0
	408	554	1294	103	41.0	14	0.0	60	24	44.0
	408	647	1201	103	41.0	14	17.6	60	24	43.0
	408	647	1201	103	41.0	12	14.4	60	24	42.0
	408	554	1294	103	51.5	14	16.5	60	24	42.0
	408	554	1294	103	51.5	14	16.5	60	24	41.0
	408	554	1294	103	51.5	14	16.5	60	24	41.0
	408	554	1294	103	41.0	14	16.5	60	24	40.0
	408	647	1201	103	41.0	16	26.5	60	24	40.0
	408	647	1201	103	41.0	14	20.7	90	4	37.0
	408	554	1294	103	51.5	14	16.5	60	24	36.0
	408	554	1294	103	41.0	14	10.7	30	24	35.0
	408	554	1246	103	41.0	8.0	20	60	18	29.0
	408	554	1080	103	41.0	8.0	20	60	18	29.0
	408	554	1243	103	41.0	8.0	20	60	18	25.0

Table 3. Cont.

Ref.	FA (Kg/m ³)	Aggregates (kg/m ³)		Alkaline Activator (AA) (kg/m ³)		NaOH Molarity (M)	Added Water (kg/m ³)	Curing Conditions		Strength (MPa)
		Fine	Coarse	Na ₂ SiO ₃ (SS)	NaOH (SH)			Temperature (C)	Time (h)	
[127]	408	616	1232	103	41.0	14	0.0	60	24	66.8
	408	616	1232	103	41.0	8.0	0.0	60	24	55.0
	408	616	1232	103	41.0	10	7.5	60	24	52.0
	408	616	1232	103	41.0	12	14.4	60	24	51.0
	408	616	1232	103	41.0	16	26.5	60	24	48.0
	408	616	1232	103	41.0	14	20.7	60	24	45.0
	408	616	1232	103	41.0	14	10.6	60	24	35.0
	408	616	1232	103	55.4	8.0	0.0	60	24	33.0
	408	616	1232	103	41.0	14	21.3	60	24	32.0
	408	616	1232	103	48.0	14	0.0	60	24	28.0
[94]	350	645	1200	103	41.0	8.0	35	80	24	20.0
[149]	420	750	1125	100.0	40.0	16	0.0	100	24	70.5
	400	535	1356	128.6	51.5	16	12.7	100	24	52.0
	380.	540	1233	141.3	56.5	16	14.6	100	24	49.0
	405	545	1235	132.4	52.9	16	28.0	100	24	46.0
	400	540	1265	105.7	42.3	16	24.3	100	24	44.0
	309	648	1204	59.0	27.7	10	83.7	100	24	42.0
	254	694	1290	48.5	22.8	10	68.7	100	24	36.8
	365	602	1118	73.0	34.3	10	103.5	100	24	35.3
[150]	408	554	1294	103	41.0	14	22.5	60	24	36.0
	350	645	1200	103	41.0	8.0	35	60	24	48.0
[134]	400	850	950	144	57.0	12	48	60	24–96	48.5
	408	554	1294	103	41.0	8.0	0.0	60	24–96	56.0
[151]	428	623	1177	102.9	68.6	14	28.5	20–23	72	28.6
[152]	400	651	1209	114.3	45.7	12	0.0	60–90	24–96	26.0
[125]	406	643	1194	102.0	41.0	14	26.8	70	12	37.0
	424	623	1177	91.0	36.4	14	16.0	70	12	54.9
	462	599	1153	132.2	52.9	14	21.2	75	24	49.6
	461	623	1177	92.3	46.2	14	18.6	75	24	42.5
	498	599	1153	89.7	59.8	14	26.5	60	24	39.9
	444	623	1177	111.1	44.4	14	18.6	60	24	38.7
	480	599	1153	112.0	56.0	14	23.7	70	12	37.1
	408	623	1177	85.9	57.2	14	24.5	75	24	35.7
	394	647	1201	105.1	52.6	14	21.5	60	24	29.7
	408	554	1294	103	41.0	16	22.5	60	24	45.0
[153]	404	640	1195	102	41.0	16	20	60	24	50.0
[154]	408	647	1201	93	62.0	14	4.0	60	4–96	32.0
	408	554	1294	103	41.0	8.0	0.0	60	4–96	58.0

7. Compressive Strength

FA-GP concrete CS is affected by S/A ratios, alkali sols, calcium content, additives, and curing conditions.

The dosage and type of alkaline sol affect the generation of Si^{4+} and Al^{3+} from FA during geopolymerization. A dosage of alkaline sol is typically advantageous for achieving high CS [3,142,155–165]; however, there is an ideal limit [166]. For instance, by raising the NaOH content from 4.5 to 16.5 M, Somna, Jaturapitakkul, Kajitvichyanukul, and Chindaprasirt [157] looked into the CS of ground FA (GFA) that had been treated at room temperature. The results showed that increasing the NaOH content from 4.5 to 9.5 M improved the CS of paste specimens. While altering the NaOH dosage levels from 9.5 to 14 M improves the CS of paste specimens, it does so to a much lesser extent. Increased alumina and silica extraction is primarily responsible for the increase in CS with increasing NaOH dosages. The CS of GFA hardened pastes begins to decline at a dosage of 16.5 M sodium hydroxide. This decrease in CS is mostly caused by excess hydroxide ions, which precipitated alumino-silicate gel at a relatively young age, resulting in the formation of weaker GPs.

To boost CS, Na_2SiO_3 sol is generally combined with NaOH [167]. This is because of the fact that Na_2SiO_3 with a high viscosity can aid in the production of GP gels, resulting in a compact final FA-GP concrete Ms. Furthermore, the activation technique affects the CS of FA-GP concrete. For instance, Rattanasak and Chindaprasirt [168] first introduced NaOH sol to FA for 10 min to dissolve the Si^{4+} and Al^{3+} species, followed by Na_2SiO_3 for one minute to assist in generating a homogeneous GP paste. The strength of FA-GP concrete increased as a result of the independent activation. Table 3 shows the specifics of the various alkaline sols as well as the molarity of the FA-GP concrete.

The FA-GP concrete structure is mostly determined by the S/A ratio generated by the dissolving of Al^{3+} and Si^{4+} . Furthermore, the efficient S/A ratios have a substantial impact on the dissolving, hydrolysis, and condensation reactions of GPs. Condensation action in a high S/A system leads to the predominance of the silicate species itself, generating oligomeric silicates, which condense with $\text{Al}(\text{OH})_4$ and create GP structures of poly(sialate-disiloxo) ($\text{Si-O-Al-O-Si-O-Si-O-}$) and poly(sialate-siloxo) (Si-O-Al-O-Si-O-) as shown in Figure 15 [169,170]. As a result, a completely dense GP structural matrix has greater CS. Using a larger dosage of an alkaline activator, in contrast, can assist rapidly in the polycondensation of alumino-silicate oligomers by the transfer of Al^{3+} and Si^{4+} [90,171–174].

The function of additives is to create a crucial equilibrium in oxide dosages to the required level using various add-ons [175–178]. Binary, ternary, and quaternary GPs have been created to achieve higher characteristics and erase any defects related to any original source [179]. For instance, Yang, Yao, Zhang, and Wang [180] found that the high calcium dosage of GBFS in FA/GBFS GP increased the initial setting time and slowed the degree of polymerization. It has been discovered that increasing GGBFS as a partial substitute for FA considerably improves both 28-day strength and short-term strength [143]. Because of the presence of C-S-H gel, iron slags as a substitute for FA in FA-GP concrete demonstrated enhanced strength with increasing slag percentage [181–183]. Kusbiantoro, Nuruddin, Shafiq, and Qazi [150] showed that samples cured in an oven had the maximum CS because of the polarization of OH ions, which tends to increase the pH of the sol and thus promotes the synthesis of -Si-O-Si- bonds in the GP gel (N-A-S-H gel) and CS [184–187]. While analyzing nano-structural changes in several GP composites (GBFS-based, FA-based, and mixed GBFS + FA) before and after carbonation, Bernal, Provis, Walkley, San Nicolas, Gehman, Brice, Kilcullen, Duxson, and van Deventer [188] revealed that N-A-S-H gel is the main product in FA-GP concrete that is not dissolved by carbonation and instead only alkali aluminosilicate (AAS) in the pores is impacted by CO_2 . The use of nano-alumina and nano-silica as additives in FA-GP has been found to improve its mechanical characteristics. Phoo-ngernkham, Chindaprasirt, Sata, Hanjitsuwan, and Hatanaka [176] created FA-GP pastes by adding 1% to 3% nano-silica and nano-alumina as an ingredient. The results

indicated that nanoparticle additions, independent of dose, enhanced CS. This behavior was attributed to both the increased formation of C-S-H or C-A-S-H and N-A-S-H gels in the GP paste and the micro packing effect. Table 4 illustrates the effect of various additives on the behavior of GP composites. Because the dissolution and condensation of FA comprise both alkali and alkali metals, the pH of the pore sol and the alkali-metal cations are crucial for the hydration process of FA and consequently the CS. Bhagath Singh and Subramaniam [189] and Yang and Gupta [190] found that the CS of FA-GP concrete rose as alkali dosage rose, and they ascribed this to the rapid dissolution of FA at higher pH. Because network modifiers (Ca^{2+} , Na^{+} , and K^{+}) are present, reacting phases including Al and Si have disorganized structures and can generate acidic -Si-OH (silanol) groups with water. The dissolution behavior of vitreous aluminum silicate-containing network modifiers is depicted in Figure 16. The presence of hydroxyl ions (OH^{-}) in alkali sols causes the reactive aluminosilicates of FA to disintegrate. The dissolution rates of aluminosilicates are affected by their exterior and interior surface areas, as well as the degree of cross-linking in their structure [191].

Calcium has been shown to interact with silica and alumina gelation during the geopolymerization reaction, altering the Ms of FA-GP concrete and hence the CS [192,193]. The presence of C-S-H gel and N-A-S-H gel together typically increases the CS of final products. One of the explanations for this is that the crystalline C-S-H gel reduces porosity [194,195]. Figure 17a depicts the fluctuation in FA-GP concrete CS after 28 days in relation to the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ (SS/SH) ratio dosage in the FA precursor.

Figure 17b depicts the change in 28-day CS of FA-GP concrete vs. CaO dosage in the FA precursor.

It is well established that typical GPs require thermal treatment to produce CSs equivalent to or greater than those of PC concrete [39,101,196–198]. Heat treatment promotes aluminosilicate gel dissolution and geopolymerization, resulting in a considerable increase in early strength [199]. Additionally, it accelerates the dissolution of silica and alumina species and the subsequent polycondensation phase. However, the thermal curing regime adopted must be suitable for promoting the proper dissolution and precipitation of dissolved silica and alumina species. Depending on the source, geopolymerization may be harmed by exceeding a specified temperature and heat treatment period, which may negatively affect the GP concrete's mechanical properties. [156,196,200]. Noushini, Castel, Aldred, and Rawal [144] used 12 different thermal curing techniques to make FA-GP concrete with a low calcium content. Figure 18 depicts the CS of GP concrete specimens after 28 days at room temperature and after heat curing. Compressive strength initially increased faster with increasing temperature and time in cured samples, and the optimal CS was obtained at 75 °C [144]. This could be because higher temperatures and longer curing times resulted in the formation of more reactive species; hence, with the appropriate curing temperature, more metals were incorporated into the GP matrix, and metal dosages decreased [201–203]. Table 5 illustrates the effect of various curing regimes on the performance of GP composites.

The type and fineness of the GP raw material are instrumental in developing the strength, durability, and Ms of the resulting GP matrices. Figure 19 depicts an SEM image of a polished part of an FA-GP with various FA particle types. Numerous researchers have concurred that altering the particle size distribution in the composite material has a substantial effect on the CS, physical properties, and Ms of the resulting GP paste [196,204]. In general, binder phases with a finer particle size distribution have a stronger reaction and, as a result, create GP paste with a denser Ms, better CS, and improved physical properties [200,205]. Table 6 shows how particle size distribution affects the performance of GP composites.

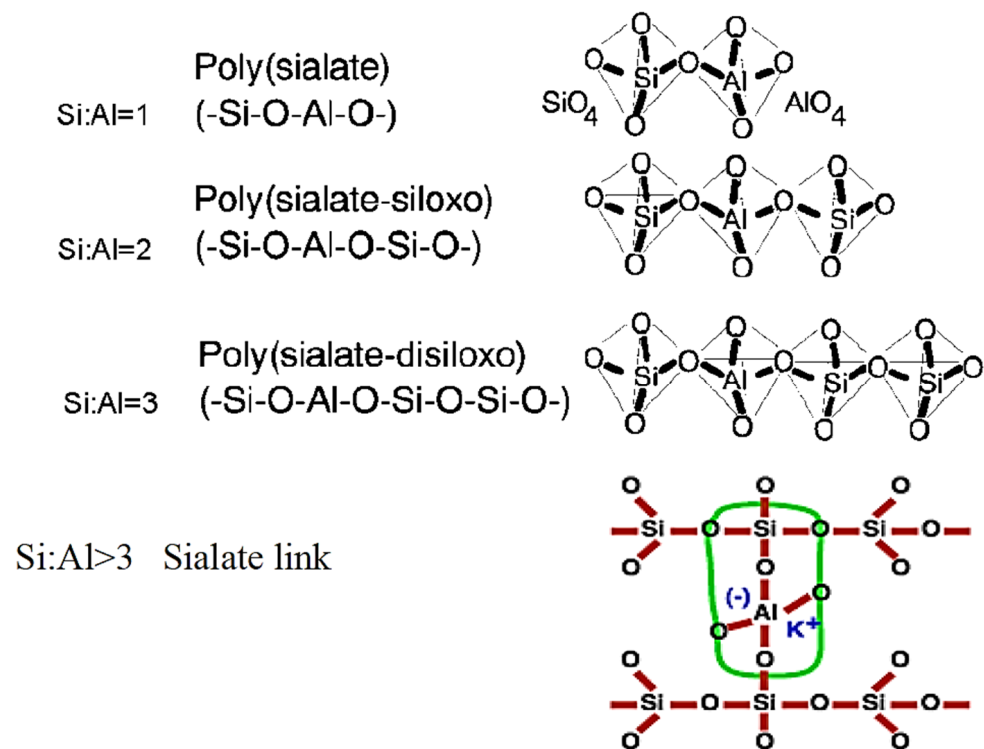


Figure 15. Formation of different aluminum silicate chains in aluminosilicate oligomers depending on the molar S/A ratio, which then forms the GP. Adapted from [85].

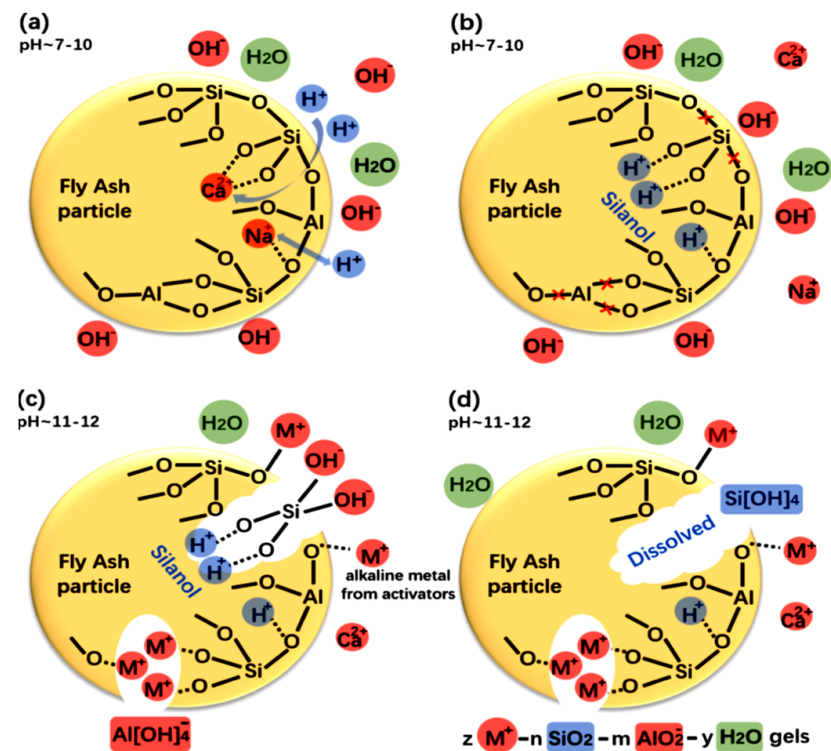
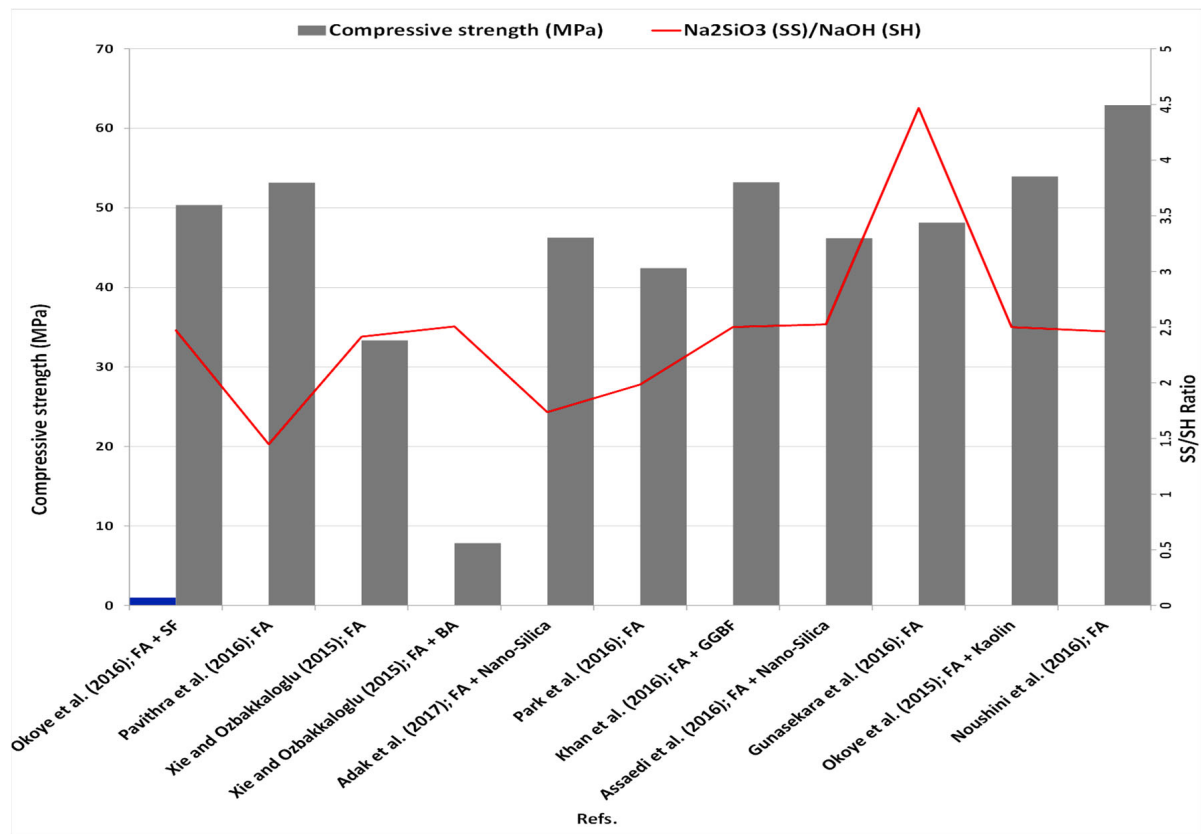
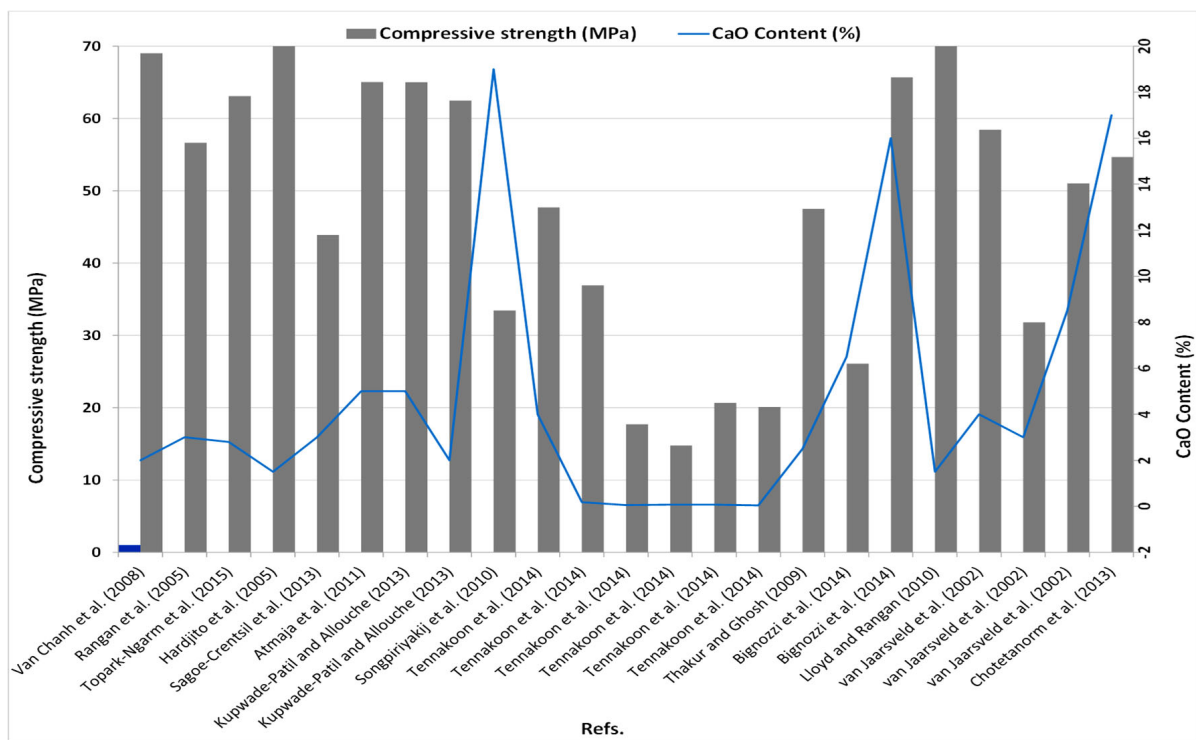


Figure 16. The process of aluminosilicate glass dissolving during the initial stages of reactivity of a one-part GP blend. (a) H^{+} exchange for Ca^{2+} and Na^{+} , (b) dissolution of $\text{Al}-\text{O}-\text{Si}$ bonds, (c) depolymerization of the glass network, and (d) Si and Al release. Adapted from [206,207].



(a)



(b)

Figure 17. Historical statistics on CS variations in comparison to: (a) Na₂SiO₃ (SS)/NaOH (SH) [93,95,96,99,103–105,107,208] and (b) CaO content in FA-GP [123,209–221].

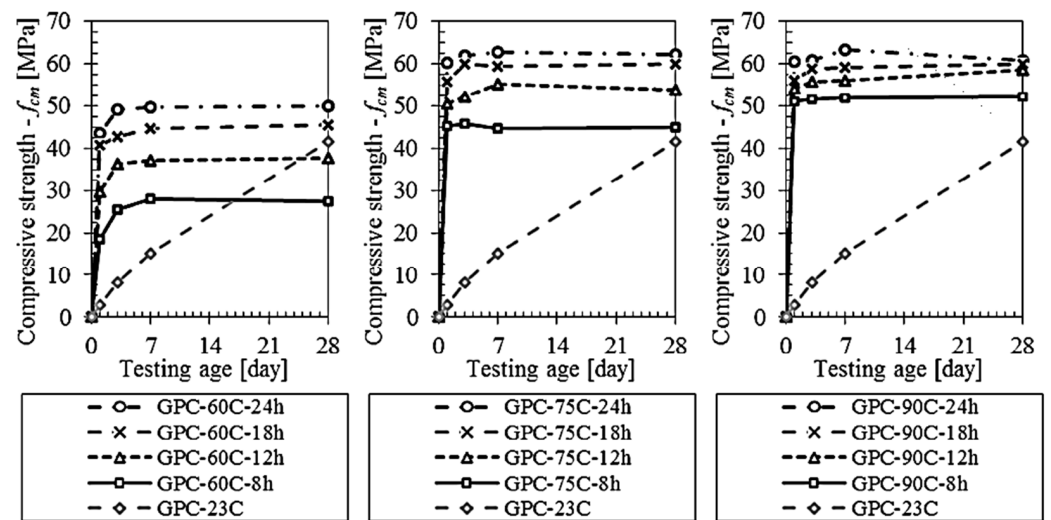


Figure 18. Compressive strength development of ambient and heat-cured GP concretes. Adapted from [144].

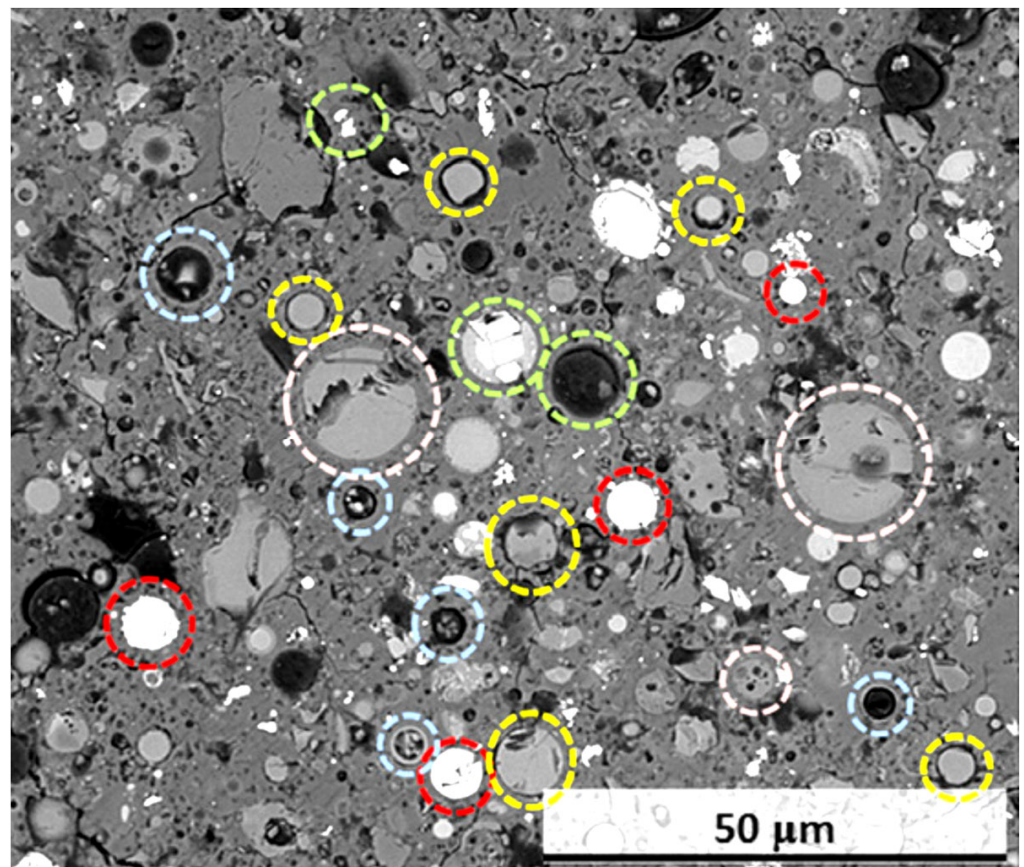


Figure 19. SEM image of a refined segment of an FA-GP concrete with several FA particle types highlighted in various colors. Yellow particles are reactive glass particles surrounded by a reaction rim [222–224]; red particles are iron-rich; pink particles are relatively unreactive glass particles with no visible reaction rim; blue particles are hollow particles whose shell was not broken during the reaction process; green particles contain visible crystalline inclusions. Adapted from [225].

Table 4. Effect of different additives on FA-GP concrete CS.

Ref.	Main Source Material	Additives	Remarks
[226]	FA	CAC	Increases strength by providing an additional supply of Al and Ca.
[227]	FA	Water glass	Increased CS because of higher Si dosage. The recommended dose is 15 g/100 mL.
[228]	FA	GBFS	With an M ratio of 0.96 SiO ₂ /Na ₂ O and raw materials of 70% GGBFS and 30% FA, CSs of 48 MPa were attained.
[226]	FA	GBFS	Compressive strength and Ms improve when GBFS dosage increases resulting from the production of more C-S-H.
[229]	High-calcium FA	PC	A GP composite with a more homogeneous and dense structure than concrete was created.
[197]	Low-calcium FA	GGBF, POFA	The addition of 30% POFA and 70% GGBS to FA-GP concrete produced a CS of 67 MPa. POFA dosages above 33% reduce CS.
[230]	FA	Superplasticizer	A high superplasticizer dose increases CS.
[231]	FA	Chitosan biopolymer	The introduction of N-carboxymethyl chitosan substantially improved tensile strength and resulted in a minor increase in CS.
[180]	FA	GGBFS	Slag inclusion in the raw material can improve the CS of GP mixtures, with a slag/FA dosage ratio of 0.8, providing the maximum strength.
[232]	FA	Sulfate of calcium and sodium, calcium chloride, and sucrose	Sucrose substantially slows down the ultimate setting time. All admixtures help to improve CS.
[233]	FA	GBFS, GCS	GCS partial replacement resulted in higher CS than GBFS partial replacement.
[234]	FA	Red mud	After 120 days, there is a decrease in CS. Localization of heavy metals within permitted levels.
[150]	FA	Incinerated rice husk ash	At an optimal dosage of 7%, addition resulted in an increase in compressive and bond strength.
[235]	FA	Nano-silica	The use of 6.0% nano-silica as a substitute caused improved mechanical properties.
[236]	FA	Aluminum-rich waste	A 2.5% admixture of dried powdered and calcined aluminum-rich waste gave early high strength of 34 MPa, but the CS dropped after that.

CAC, calcium aluminate cement; POFA, palm oil fuel ash; GCS, ground corex slag; GGBFS, ground granulated BFS.

Table 5. Curing regime and its influence on FA-GP concrete CS from different precursors.

Ref.	Precursor	Curing Regime	Remarks
[237]	FA	Curing temperatures between 66 °C and 85 °C	Curing at 85 °C for 24 h resulted in much greater strength than curing at 66 °C. Over time, the rate of growth in strength dropped after 24 h.
[167]	Class C FA	I: 62 °C for 24 h; II: 23 °C at room temperature.	The early strength of HCGC is influenced by higher curing temperatures. Compressive strength increases rapidly over the first 7 days and then gradually increases for the next 28 days.
[212]	FA/silica fume	I: 96 °C (8 h); II: 96 °C (2 h) then 150 °C (6 h) dry oven; III: 96 °C (2 h) then 96 °C (6 h) steam	50–102 MPa for I; 28–57 MPa for II. The greatest results were obtained with III: 35–77 MPa, covering with 95 °C, and steam curing.
[234]	FA slag	I: 26 °C (28 days); II: 26 °C (48 h) followed by 60 °C (4 h)	The second curing regime, which was longer in length and followed by high-temperature treatment, resulted in enhanced strength.

Table 5. Cont.

Ref.	Precursor	Curing Regime	Remarks
[167]	FA	85 °C (5 h–7 days)	Strength increases when curing time is extended. Sealed curing aids in the development of strength while also preventing early carbonation.
[238]	FA	26 °C (16 h–672 h); 41 °C (72 h) to 336 h; 60 °C (16 h–120 h); and 85 °C (1 h–6 h).	85 °C for 6 h is equivalent to 25 °C for 100 h. The strength of the K-based GP was reduced as the curing temperature increased.
[239]	FA	I: 21 days at 20 °C with an initial cure at 70 °C; II: 24 h at 20 °C with an initial cure at 70 °C.	Technique II has a higher CS than method I.
[220]	85% FA and 15% kaolin	6 h, 12 h, 24 h, and 48 h for temperatures of 30 °C, 50 °C, and 70 °C.	Curing at a greater temperature for a brief time has a positive influence on strength (a couple of hours). Structure deterioration with prolonged exposure to high temperatures.
[240]	FA	24 h at 65 °C; 5 min in the microwave. After 66 °C curing for 3 h, 6 h, and 12 h, curing at room temp	The optimal curing time was 5 min in a microwave at 65 °C for 6 h.
[241]	FA	1 h of pre-curing followed by 24 h of curing at 25, 40, and 60 °C.	Curing at 60 °C is ideal (for 7- and 28-day strength)
[200]	FA	24 h, followed by 36 h in the oven (50–90 °C).	Curing in an oven at 80 °C is ideal.
[242]	FA	Room temperature for 9–12 h, then salt water, normal water, and sealed curing environment.	The sealed state produced the best outcomes, followed by salt water and the least effective water cure.

HCGC, high calcium FA-GP concrete.

Table 6. Impact of the particle size distribution of precursors on FA-GP concrete CS.

Ref.	Precursors	Precursor Gradation	Main Constants	Compressive Strength (MPa)	Remarks
[243]	FA	100% GFA (1.45 m ² /g)	Na ₂ SiO ₃ with a silicate modulus of 2.5.	6.79	With the addition of finer binder content, the CS of 28 days is increased.
		25% OFA (0.395 m ² /g) + 75% GFA		4.65	
		50% GFA and 50% OFA		3.85	
		25% GFA and 75% OFA		3.84	
[244]	FA	365 m ² /kg	NaOH dosage of 13 M, Na ₂ SiO ₃ /NaOH ratio of 3, Sol/FA ratio of 0.35, curing time of 7 h at 110 °C	21.85	With increasing fineness, there is a substantial rise in CS after 28 days.
		440 m ² /kg		29.7	
		612 m ² /kg		42.29	
		d ₅₀ = 32.24 µm and fineness		40	
[245]	FA	2110 cm ² /g	Alkali sol with a dosage of 13 M was utilized, and the L/S ratio was 0.35 after 4 h of curing at 90 °C.	62	With an increase in fineness, CS and workability improve after 28 days.
		525 m ² /kg		57.62	
		444 m ² /kg		39.22	
		365 m ² /kg		9.5	
[246]	FA	X ₁₀ = 0.12 µm, X ₉₉ = 3.66 µm,	Na ₂ SiO ₃ /NaOH ratio of 0.4 and activator to FA ratio of 0.45%.	15.5	With an increase in fineness, there is a substantial rise in the CS of mortar after 28 days.
		X ₁₀ = 0.08 µm, X ₉₉ = 0.64 µm,		19.3	
		X ₁₀ = 0.04 µm, X ₉₉ = 0.37 µm,		34	
[200]	FA, RHBA	FA 75 µm + RHBA 90 µm	Curing at 80 °C for 36 h with a NaOH dosage of 12 M.	44	With an increase in fineness, there is a substantial rise in the 28-day CS of mortar.
		FA 75 µm + RHBA 7 µm		52.6	
		FA 3 µm + RHBA 90 µm		59.1	
		FA 3 µm + RHBA 7 µm			

Table 6. Cont.

Ref.	Precursors	Precursor Gradation	Main Constants	Compressive Strength (MPa)	Remarks
[205]	FA, RHA	5.1% retained on sieve 325 of RHA 1.1% retained on sieve 325 of RHA	As-received FA, $\text{Na}_2\text{SO}_3/\text{NaOH}$ ratio = 4, 60 °C for 48 h.	34.5 43	Increased fineness resulted in an overall enhancement in CS after 28 days.
[204]	BA	$d_{50} = 15.8\mu\text{m}$ and fineness 5100 cm^2/g $d_{50} = 24.4\mu\text{m}$ and fineness 3500 cm^2/g	Curing at 75 °C for 48 h with a $\text{Na}_2\text{SO}_3/\text{NaOH}$ ratio of 1.5 and a NaOH dosage of 10 M.	64.5 48	With a rise in fineness, the CS of mortar increases in 28 days.
[158]	BA	Particle size of 15.7 μm	Liquid alkaline/ash ratios of 0.429–0.709, $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratios of 0.67–1.5, and 7.5–12.5 M NaOH.	24–58	Grinding boosts reactivity and lowers porosity in BA particles, resulting in reasonably high workability and CS of 28 days.

GFA, ground FA; OFA, oil FA; RHA, rice husk ash; RHBA, rice husk-bark ash; BA, bottom ash.

8. Microstructure (Ms)

8.1. Scanning Electron Microscopy (SEM)

The Ms characteristics of FA-GP concrete rely on the FA characteristics, type, and dosage of activators and the curing conditions [95,96,98]. Fernández-Jiménez, Palomo, and Criado [247] presented an explanatory model of FA-GP concrete as presented in Figure 20. Before defining the alkaline activator sol, Pavithra, Srinivasula Reddy, Dinakar, Hanumantha Rao, Satpathy, and Mohanty [95] established several fixed parameters, including dosage, SS/SH ratio, curing temperature, and duration (AAS). This study discovered that as the ratio of AAS to binder increases, the strength of GP concrete decreases because of the increased amount of water in the GP mix [16]. The contact area for the reaction was obstructed by the water molecules, and this, in turn, altered the geopolymerization process between the binder and the activator, resulting in a low compressive strength of the GP concrete [248]. When AAS/binder ratios of 0.4 and 0.5 were employed, the GP concrete matrix included a negligible percentage of unreacted FA particles. Nonetheless, it has been demonstrated that the presence of unreacted FA particles in the FOC matrix aids in matrix densification and minimizes the formation of microcracks. This behavior is consistent with other studies demonstrating that the ability of unreacted FA particles to densify the Ms of GP concrete improves strength performance [96,98,249,250].

The influence of several SS to SH ratios on Ms studies has been investigated [251]. As shown in Figure 21, fully reacted FA particles were evident in GP concrete when an SS to SH ratio in the range of 0.5 to 1.0 was used. Despite the similarity in the Ms of GP concrete made with SS to SH ratios of 0.5 and 1.0, the resulting CS differs. The CS of GP concrete made with an SS to SH ratio of 1 was 65 MPa, while the GP concrete made with an SS to SH ratio of 0.5 exhibited a CS of 35 MPa for the same age. These strength findings are in agreement with a previous study where a higher dosage of silicate was found to result in better strength development as a result of the availability of additional silicates available for geopolymerization [237]. However, the Ms images presented in Figure 21 show that GP concretes made with a higher SS to SH ratio have more presence of unreacted FA particles. It is well established that an excess of AAS in the mix hampers geopolymerization by restricting the interaction of the binder and AAS [252]. When current research on this subject is compared, it is clear that most writers utilized a mass ratio of SS/SH of up to 2.5 [93,96,208,253].

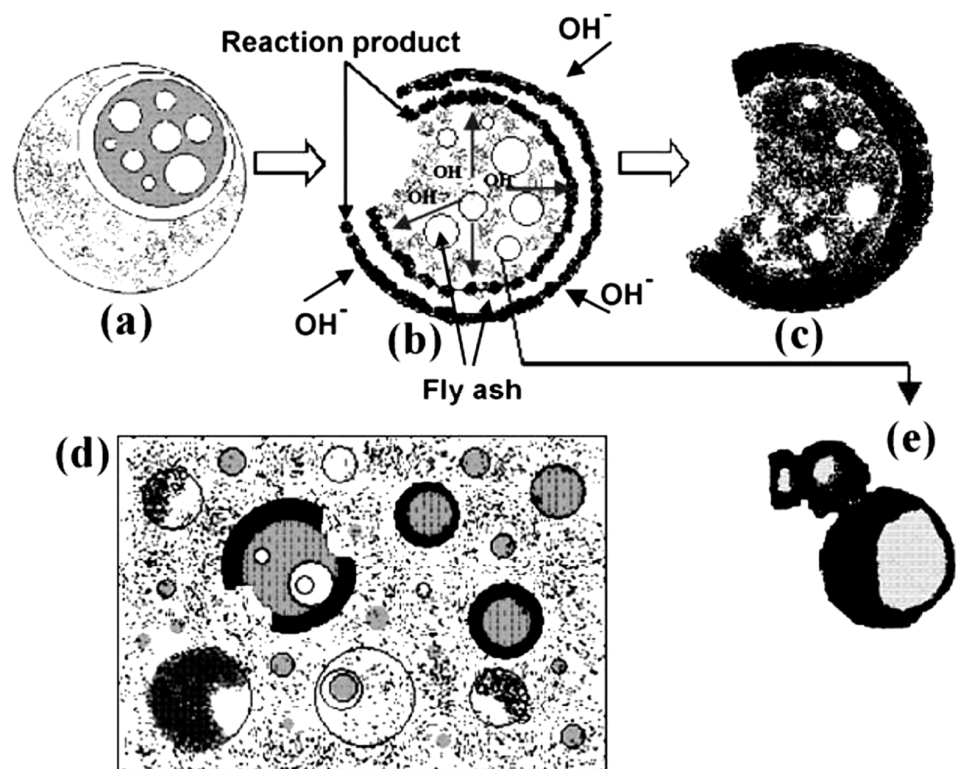


Figure 20. A descriptive model of FA activation by alkali: (a) initial chemical attack, (b) bi-directional alkaline attack, (c) reaction product, (d) gel, (e) unreacted particles. Adapted from [247].

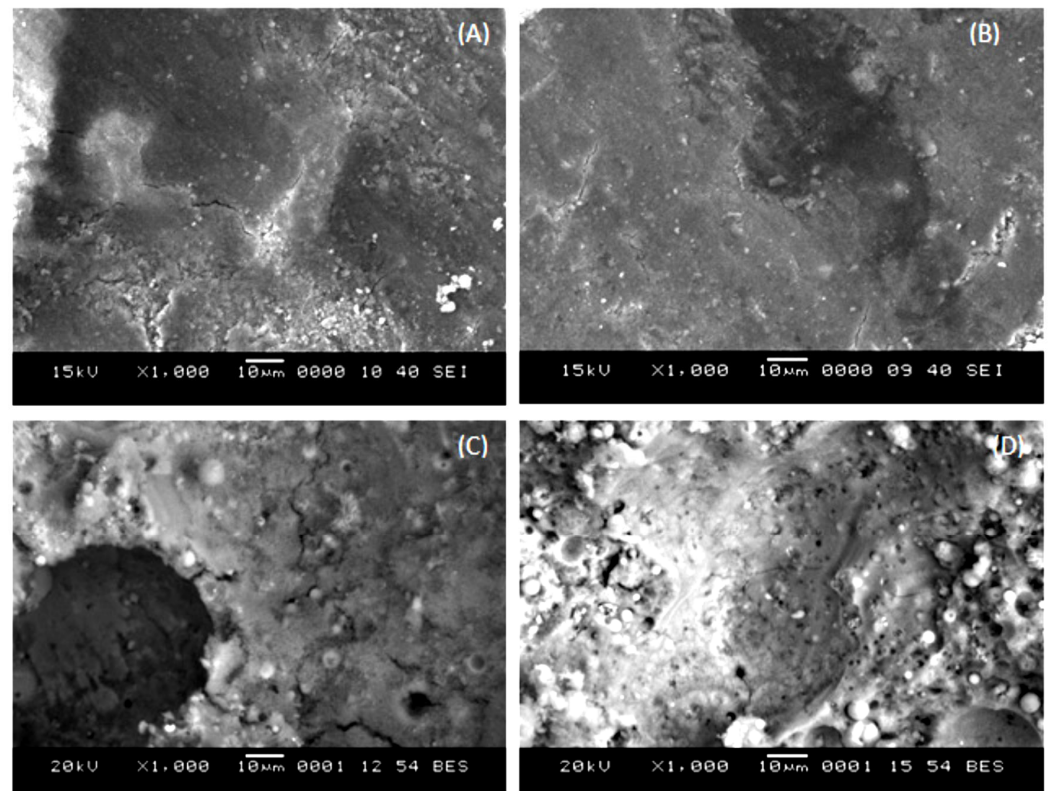


Figure 21. SEM images of FA-GP concrete generated at various $\text{Na}_2\text{SiO}_3/\text{NaOH}$ weight proportions at 28 days: (A) 0.5, (B) 1, (C) 2, and (D) 3. Adapted from [251].

Karthik, Sudalaimani, Vijayakumar, and Saravanakumar [254] included bio-additives in GP concrete and established a correlation between the Ms density and CS. It was discovered that increasing the density of the GP concrete Ms results in increased CS. These findings corroborate previous research on including nano-silica, silica fume, nano-aluminate, and graphene oxide in FA-GP concrete [255,256]. Figure 22 shows SEM images of GP concrete nanocomposites containing 3.0 wt% nano-silica in wet and dry mix conditions. The amount of unreacted particles has been substantially reduced. Additionally, the dry mix process detects fewer microcracks than the pure GP and wet mix procedures. Further, Cai, Pan, Li, Tan, and Li [257] report that FA-GP concrete has a higher electrical resistivity than metakaolin-based GP concrete.

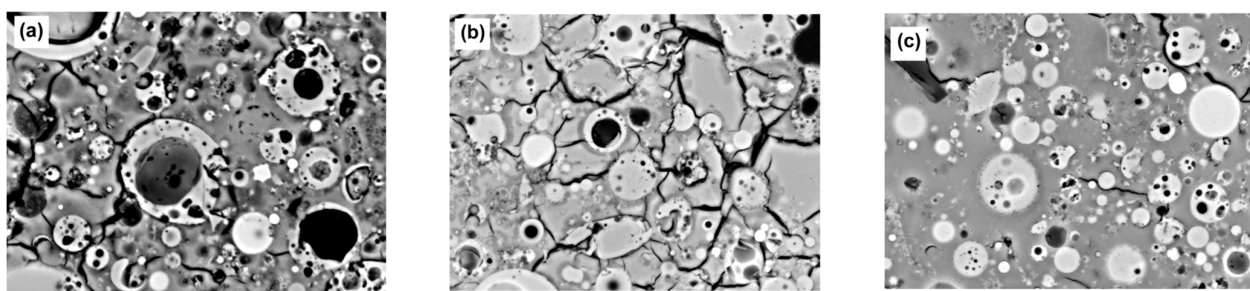


Figure 22. SEM images showing (a) pure GP combined with GP nanocomposites comprising 3.0 wt% nano-silica, (b) wet mix procedure, and (c) dry mix procedure. Adapted from [104].

Figure 23 shows the interfacial transition zone (ITZ) in GP concrete. According to Embong, Kusbiantoro, Shafiq, and Nuruddin [100], the enhancement of FA particle dissolution and polycondensation of alumino-silicate compounds provides a link between the aggregates and the matrix. The CS of the samples was greatly increased as a result of the development of alumino-silicate filling the ITZ.

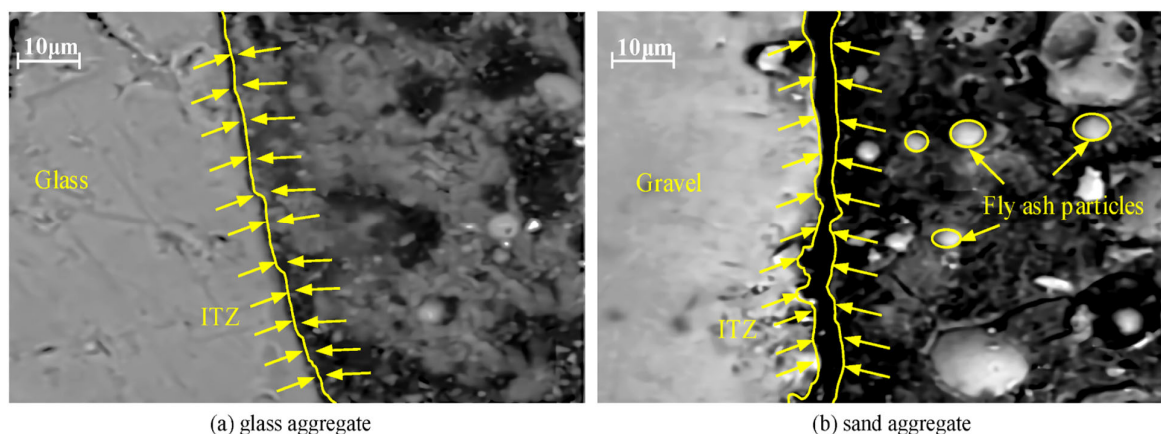


Figure 23. FA-GP concrete SEM images with several types of aggregates: (a) the interface of the glass aggregate and GP; (b) the interface of the sand aggregate and GP. Adapted from [258,259].

8.2. Wide-Angle X-ray Properties

In reality, Lloyd, Provis, and van Deventer [260] chronicled that the boost in the quantity of silica encourages the diffusion of alkalis. Nevertheless, these phase modifications taking place during aging are absent in the XRD patterns. In addition, Abdulkareem, Mustafa Al Bakri, Kamarudin, Khairul Nizar, and Saif [97] described a broad hump ranging from 20° to 35° that indicated the presence of amorphous GP products. Previous research has revealed that FA comprises quartz and mullite mineral phases, and Table 2 also shows that iron and calcium made up more than half of the FA [261]. As a result, the reason for the presence of phases in the GP paste pattern is depicted in Figure 24. Mullite $\text{Al}_{4.56}\text{Si}_{1.44}\text{O}_{9.72}$,

quartz SiO_2 , magnetite Fe_3O_4 , and calcium silicate Ca_3SiO_5 were recognized as crystalline phases. The inclusion of calcium silicate in the quantitative measurement of fly ash indicates slight cement contamination in Melbourne ash. The binder's XRD phases are entirely connected to the fly ash source material, and no novel crystalline phases are generated in this binder. Furthermore, the amount of the calcium silicate phase does not decrease with time, indicating that the C3S phase in fly ash does not participate much in the reaction, and the sodium aluminosilicate hydrate (N-A-S-H) gel is still the predominant binding matrix in these specimens. As previously discussed, crystalline phases were revealed after adding an alkaline sol.

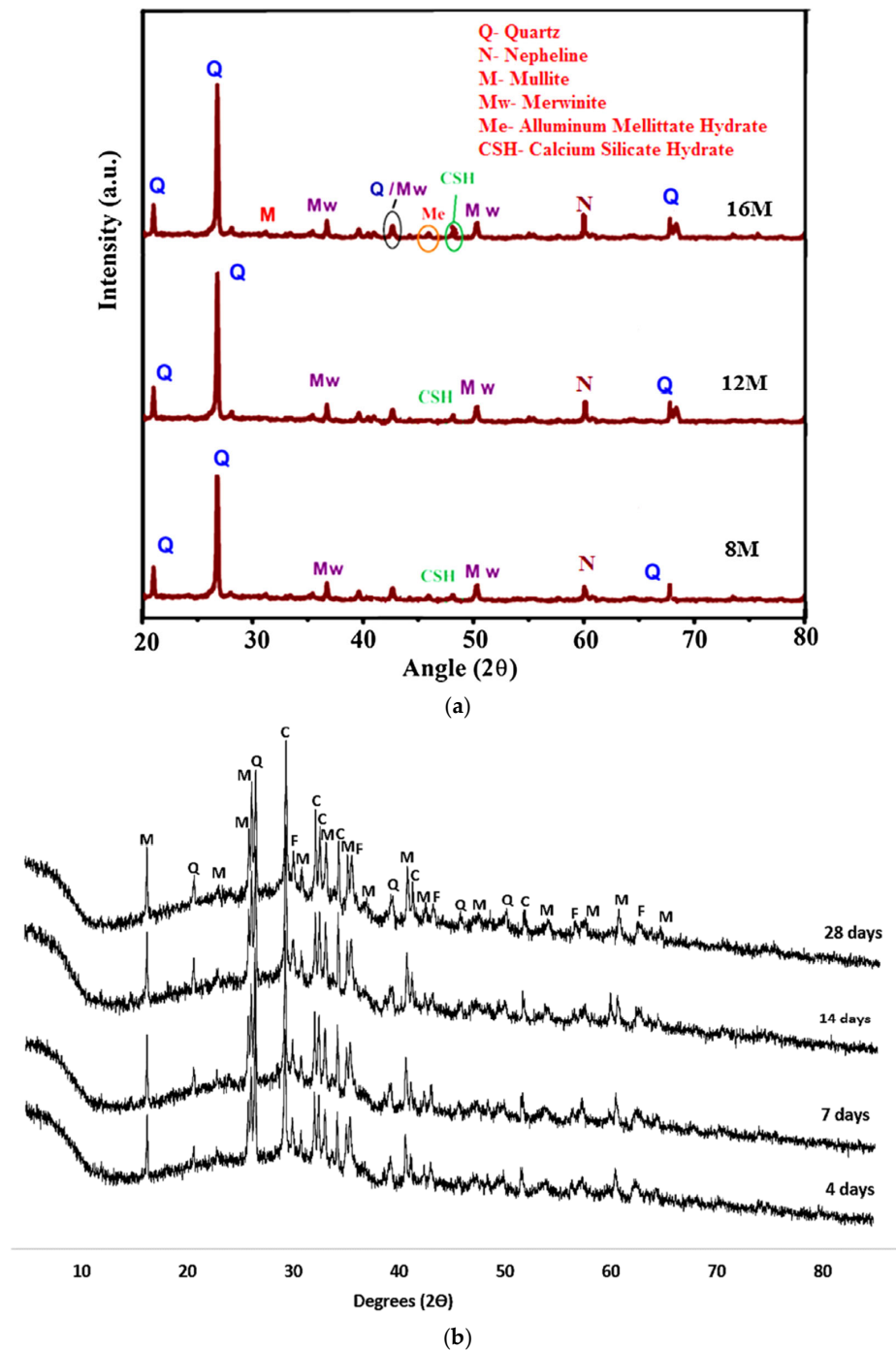


Figure 24. XRD pattern of FA-GP concrete specimen at different (a) molarities after 28 days and (b) ages. Adapted from [258,262].

As shown in Figure 25, the intensity of most peaks on the XRD spectrum is weaker and a considerable amount of amorphous phases still exist after the dissolving testing. This is because the calcium content in the FA can produce fast precipitant (Portlandite) covering the surface of the FA, which not only prevents further dissolution of the FA but also reduces the intensity of the peaks of the crystalline phases [108].

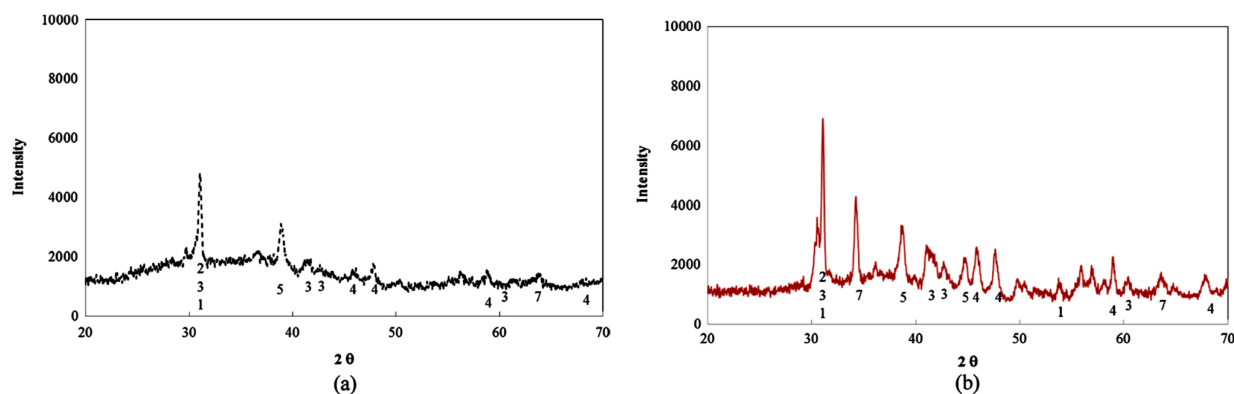


Figure 25. XRD spectrum (a) before and (b) after the dissolution of FA. Adapted from [108].

8.3. Mercury Intrusion Porosimetry (MIP)

The study by Aligizaki [263] showed that porosity can assist in understanding the characteristics of GP concrete and PC as it is a sign of the density of the Ms, microcrack presence, and diffusion rate of pore sol. Das, Yang, Singh, Mertens, Xiao, Chawla, and Neithalath [110] examined the dimension of the pore structure of FA-GP concrete in the range of 0.0036–10 μm MIP. As shown in Figure 26, the cumulative porosity for FA-GP concrete is 32%. However, the majority of the pores measured in the GP matrix were 0.0036–1 μm in size.

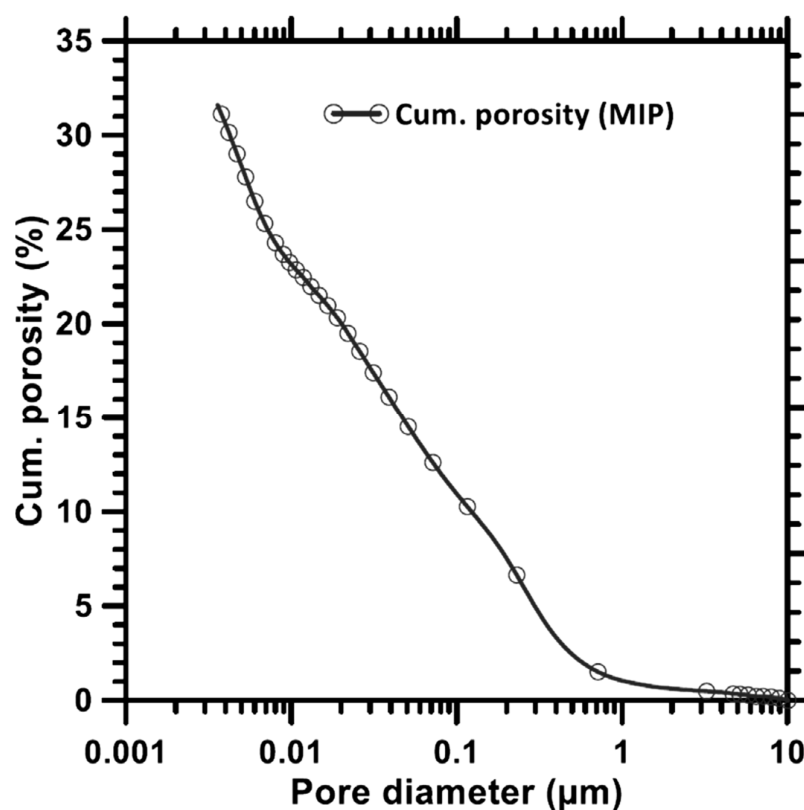


Figure 26. Cumulative mercury intrusion curve for alkali-activated FA. Adapted from [110].

8.4. GP Concrete and PC Ms Comparison

As demonstrated in Figure 27, hardened GP concrete has a denser Ms than hardened PC because of the previously noted cross-linked characteristic. Because of the replacement of Al^{+3} for Si^{+4} in bridging locations, the C-A-S-H matrix chains in the GP system were shown to be longer than the C-S-H gel chains in the PC system [35]. Because of the stronger interlayer cohesion caused by the bonding between bridging tetrahedral conservative layers, C-A-S-H gels have a lower Ca/Si ratio and a higher Al/Si ratio than C-S-H gels [264]. The gel composition of GP concrete and PC is shown in Figure 28.

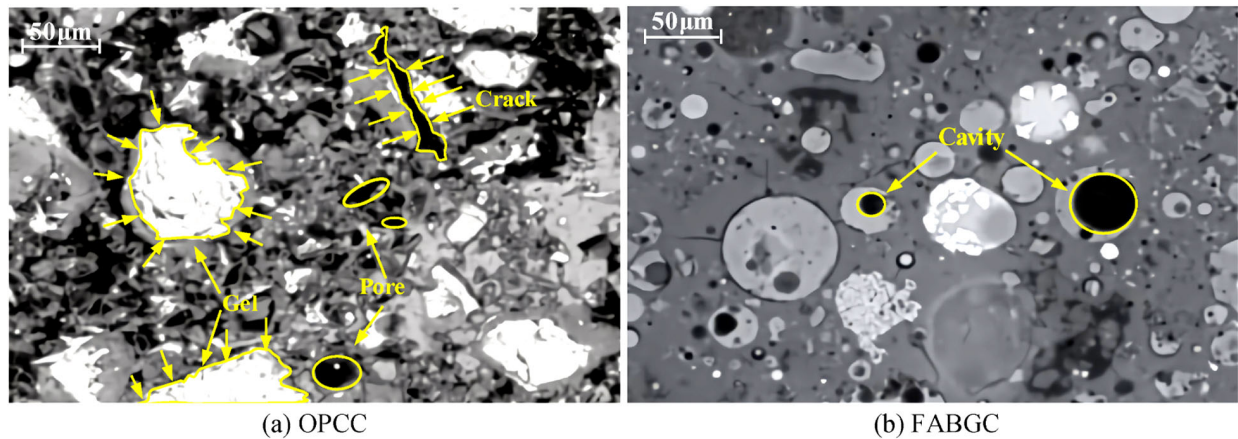


Figure 27. SEM images of the binders and ITZs of (a) PCC and (b) FA-GP concrete. Adapted from [172,259,265].

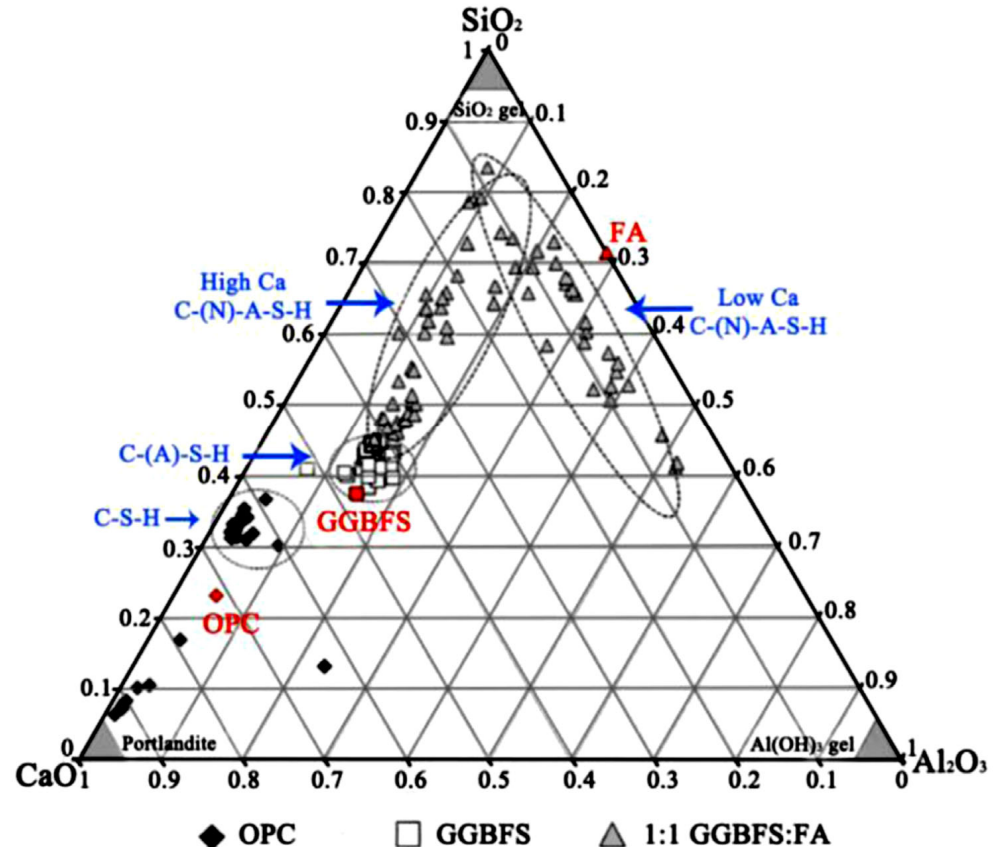


Figure 28. GP and PC binder gel compositions. Adapted from [66].

9. Challenges for Solid Wastes in Geopolymer

To accomplish the industrialization and commercialization of geopolymers, hurdles must be overcome in three areas: technology, economy, and administration. This also necessitates collaboration between the government, businesses, and the general people for co-governance, which is both an objective necessity for enhancing the quality of the ecological environment and a practical prerequisite for constructing an ecological civilization. Geopolymers derived from solid wastes are expected to become more frequently employed when nonrenewable resources are depleted. Basic research should be prioritized in order to minimize the cost and increase the workability of geopolymers. Based on fundamental research, embarking on solid wastes to explore the preparation of varied characteristics of geopolymers in order to produce high-value-added application domains should be one of the primary paths of future geopolymer research development [1,266–270].

10. Conclusions

With the main alumino-silicate precursors used in the production of GP concrete being industrial wastes, such as FA, the utilization of GP concrete in place of the conventional PCC can be used to reduce CO₂ emissions. The use of GP concrete would also result in an effective way to manage industrial wastes such as FA, thereby promoting the sustainable development of society. Discussions presented in this review paper showed that FA-GP concrete could be an alternative to PCC. Nevertheless, this will only happen if both an efficient raw material supply chain and a product supply chain network are in situ. From this perspective, the recent market is optimistic, but it will take time to establish GP concrete as a commercial product worldwide. Desirable performance for diverse engineering applications may be achieved with correct selection and adjustment of GP concrete composition. Regardless of the differences in the precursors' characteristics, the production and formation and the characteristics of the FA-GP concrete products are highly influenced by the physical and chemical characteristics of FA, AAs, additives, and curing conditions. Based on the discussion in this paper, the following conclusions can be made:

1. The recommended S/A ratio for producing GPs of appropriate strength varies depending on the nature and content of the raw material. S/A ratios of 2–2.5 (class F FA) and 2.5–3.5 (class C FA) were determined to be optimal. The influence of Al and Si on setting and hardening characteristics in high-Ca FA is substantial.
2. The conventional AAs, a combination of NaOH and Na₂SiO₃, are very good and economically viable alkali activating sols. Microstructural investigations revealed that GP concrete prepared with a lower NaOH to Na₂SiO₃ ratio reacts more efficiently because of the large surface area available for reaction and binding without being clogged by excess water. However, the hydroxyl groups tend to condense when specimens are heated to high temperatures.
3. FA-ITZ GP concrete consists of gel, pores, fissures, and unreacted FA particles. N-A-S-H gel can improve the strength of the ITZ by promoting the combination of the aggregate interface and the GP matrix. As a result, FA-GP concrete has nearly no obvious weak ITZ near the aggregate.
4. The existence of C-S-H gel, as well as GP gel, improves the mechanical and Ms characteristics of precursors either having high Ca or mixed with Ca components.
5. When it comes to GP synthesis, the criticality of precursor oxide dosage cannot be overstated. In general, the Si oxide content should be between 45% and 55%, the Al dosage should be between 22% and 28%, and the FeCO₂ and CaO content should be between 15% and 20%.
6. The experiment using MIP analysis reveals that extending the curing duration of GP specimens is useful in minimizing pores. The increased geopolymerization limits the connection between the pores, leading to a denser pattern. Furthermore, FA particles and gel may be used to refine the pores in FA-GP concrete. As a result, FA-GP concrete gel pores and capillary pores are concentrated in the tiny pore-size region. However, it

is worth noting that the silicon content of the AA plays an essential role in the refining of the FA-GP concrete pore structure.

7. It was found that increasing the fineness leads to an increase in the reaction rate and therefore requires a minus time of heating to achieve strength, as it was found that more than 60 MPa can achieve the strength of GP concrete by increasing the fineness of the precursor material.
8. FA-GP concrete with a compact and denser structure shows high mechanical strength and good efflorescence that make it an ideal choice for the construction industry and has been effectively used in precast industries. Moreover, there is an enormous possibility of utilizing high-Ca GP as a restorative material.

11. Recommendations

Though FA-GP concrete is a relatively new, innovative, and sustainable engineered material combination with numerous advantages, some difficulties remain, and some are recommended below for further investigation:

1. Currently, the findings of relevant investigations on the Ms of FA-GP concrete are discordant in several respects. For example, the conclusions on forming a new crystalline phase prior to and during the AA reaction of FA are inconsistent. As a result, additional systematic and in-depth research is required to prove this substance's usefulness with greater certainty. Additionally, advanced analytical techniques such as micro- or nano-analysis and nuclear magnetic resonance must be employed.
2. Suitable guidelines for the selection of aggregate contents in FA-GP concrete should be developed. It is also critical to develop design procedures for each substantial predecessor. Furthermore, the usage of dune sands, fibers, marble dust, date ash, and other materials as precursors should be investigated.
3. Detailed investigations, such as those relating to the derivation and modeling of reaction kinetics under different treatment and production circumstances of the developing class of GP raw material, such as blended GPs and biomass ash GPs, are needed.
4. GP binders need a high pH and heat curing. As a result, efforts are needed to create a room-temperature-cured one-component GP system that uses solid activators rather than alkaline sols in order to gain widespread acceptability in the field.
5. FA-GP concrete should be given more functionality instead of using it as an alternative cement only. FA-GP concrete with biomass has the potential for development as a new class of lightweight fireproof composites.
6. Lastly, more research is needed to evaluate the cost of GP concrete compared to that of standard concrete. Furthermore, contradictory findings regarding the embodied energy and carbon footprint of GPs in comparison to traditional Portland cement must be addressed.

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